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Date August 1987

DEVELOPMENT OF AN ACOUSTIC LEVITATION APPARATUS FOR A COAL PYROLYSIS STUDY

A Thesis

Presented for the

Master of Science

Degree

The University of Tennessee, Knoxville

Joseph E. Marquis

August 1987

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ABSTRACT

An acoustic levitator was designed and constructed to hold coal particles stationary in air. It consisted of a simple, single axis driver-reflector system that allowed for optical focusing, observation and photography. One at a time, particles were suspended in a standing ultrasonic wave, then heated with focused thermal radiation. The main advantages of this study were being able to clearly view a known particle reacting because only single particles were used and holding that particle in position without any solid body contact when heating.

The behavior of particle heating in the levitator was observed through a microscope and noted. With heat application, particles were seen to roll out, explode out or jet out of position. The external restructuring, upon softening, of a few coal particles was magnified and captured on film. This research demonstrated the development of an acoustic levitation apparatus as a potentially improved coal pyrolysis experiment over previous experiments.

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LIST OF SYMBOLS

Symbol	Definition	Units
c	speed of sound in medium	[m/sec]
E	activation energy	[cal/mol]
FC	fixed carbon content	[%]
g	gravitational acceleration	[m/sec^2]
k	rate constant	[sec^{-1}]
k_0	empirical constant	[sec^{-1}]
k'	empirical constant	[sec^{-1}]
n	any integer	
Prms	root mean square pressure	[Pa]
R	gas constant	[cal/K mol]
T	temperature	[K]
t	time	[sec]
VM	volatile matter content	[%]
VM*	effective volatile matter content	[%]
λ	wavelength	[m]
ρ	medium density	[kg/m^3]
ρ_s	particle density	[kg/m^3]

CHAPTER I

INTRODUCTION

Coal pyrolysis is the evolution of gaseous products from coal with the application of heat. During pyrolysis, coal particles undergo major physical and chemical changes, and lose from a few percent to 80 percent of their total weight. Pyrolysis is an essential aspect in all coal conversion processes because further reactions depend on the quality and quantity of the changes.

The benefits of understanding pyrolysis have prompted many different experiments. Heating a physically restrained sample or moving particles (by a gas stream or by gravity) through a heating environment are the two major methods. In the first, there is the possibility that the particles will react differently when packed in a group than singularly and may in fact react with the container. In the second, the particles may be damaged from collection procedures. In both, there is the disadvantage of not being able to observe the particle during pyrolysis.

The approach that this research took to avoid the drawbacks of previous experimental methods was to free a coal particle from solid body contact while holding it stationary. An apparatus that provided these conditions

was the acoustic levitator. An acoustic levitator can fulfill the need to suspend a small coal particle (about 100 to 500 micron diameter) in a gas. The single-axis levitator works by producing a standing ultrasonic wave between a sound source and a reflector. A particle stays on a surface in the sound field where the pressure is at a minimum. Coal particles were heated while suspended in the levitator.

Conduction, convection and radiation modes of heating have been used in previous experimental methods. In simulating pyrolysis in combustors, it is best to use radiative heating because when a coal particle enters a coal flame, it encounters a rapidly changing radiative environment (George, 1984). To irradiate a particle up to pyrolysis temperatures in the levitator, two small lasers were aimed and two broad band lamps were focused on the particle.

This research is a study of coal particle pyrolysis using acoustic levitation. There were three objectives of this project:

1. to develop an acoustic levitation device to hold single coal particles stationary in a gas,
2. to focus optical equipment on the particle to heat it,

3. to observe by means of microphotography some coal particles, with known composition and history, throughout the pyrolysis process.

The first objective was met by designing and constructing a levitator and then refining the design. Items of equipment were purchased and available materials were used to meet the goal. To complete the second objective, the radiation equipment was organized and surrounded the levitation apparatus. Thirdly, using a 35-mm and a 16-mm high speed camera, coupled to a microscope, some details of coal external restructuring were recorded on film during heating.

In Chapter II, the literature pertaining to coal pyrolysis and acoustic levitation is reviewed. The physical description, experimental methods and mathematical modeling of coal pyrolysis is discussed. Chapter III describes in detail the technique used in this research. Chapter IV displays and discusses the results obtained. Chapter V summarizes the project then provides recommendations for additional studies.

CHAPTER II

LITERATURE REVIEW

Coal Pyrolysis

Physical Description

When coal is heated in an inert or reactive environment, it undergoes restructuring and mass loss as a result of softening and gaseous material evolution. Upon heating, coal experiences a series of complex physical and chemical changes. In this section of Chapter II, a selective description by several authors of coal pyrolysis is surveyed. The current work can then be placed in perspective with the previous work.

Coal is a heterogeneous, organic material with similarities and differences between its different types. In early literature, coals were classified as bright, predominately bright, or dull. These designations are due to coal's varied appearance: glossy, rough or fibrous (Berkowitz, 1979). Coal structure is highly planar and layered. Interwoven into these layers are air spaces and coal is sometimes referred to as a solid colloid, because it is porous with a macropore and a micropore volume of 8-20 percent (Smoot, 1979). This pore space strongly influences the reactive aspects of coal. Mass transfer through the pores during pyrolysis is discussed later.

Coal is currently ranked by qualitative measurements of volatile matter (VM), fixed carbon content (FC) and heating value. Volatile matter consists of hydrocarbons, carbon monoxide, carbon dioxide and chemically combined water. Fixed carbon is defined by

$$\%FC = 100 - (\%H_2O + \%VM + \%ash). \quad [1]$$

Low rank coals including lignites and subbituminous coals have little fixed carbon and much volatile matter. Subdivisions of low rank coals are classified by their calorific value or heat of combustion. Bituminous coals and anthracites, with much fixed carbon and little volatile matter, have a high rank. Table 1 lists coal rankings by the American Society of Testing Materials (ASTM) (Berkowitz, 1979).

Product Distribution

Weight loss during heating depends strongly on temperature and heating rate. Pyrolysis can be divided into three temperature dependent stages. In the first (low temperature) stage, there is small weight loss. Raw coal contains moisture on its surface and in the pores; the effect of initial heating is to drive the free gases and water from the coal surface and pore structure. Although most of the uncombined water is driven off below 105 °C, this water is not completely removed until the

Table 1.

ASTM coal classification by rank
(Berkowitz, 1979)

Class and group	FC (%) Dry-ash-free	VM (%) Dry-ash-free	Heating value (Btu/lb)
I. Anthracite			
1. Metaanthracite	>98	<2	
2. Anthracite	92-98	2-8	
3. Semianthracite	86-92	8-14	
II. Bituminous			
1. Low volatile	78-86	14-22	
2. Medium volatile	69-78	22-31	
3. High volatile A	<69	>31	>14,000
4. High volatile B			13,000-14,000
5. High volatile C			10,500-13,000
III. Subbituminous			
1. Subbituminous A			10,500-11,500
2. Subbituminous B			9500-10,500
3. Subbituminous C			8300-9500
IV. Lignite			
1. Lignite A			6300-8300
2. Lignite B			<6300

temperature reaches around 300 °C due to surface tension effects in the small pores.

The second stage is sometimes termed active thermal decomposition. From between 350 to 550 °C, about 75 percent of all volatile matter including all tar and lighter condensible hydrocarbons is evolved (Berkowitz, 1979). During active thermal decomposition, the coal molecules may fragment and gaseous free radical species randomly recombine at the pore walls. Chemically, a coal molecule is made up of various repeating molecular structural units or polymers. Depolymerization occurs due to the rupture of weak bridges, i.e., bonds which can be thermally broken. There is a short supply of unattached hydrogen in coal. The light hydrocarbons and other species compete with the tar for the coal's donatable hydrogen to stabilize the free radicals (Solomon and Hablen, 1985).

The volatiles evolve from a coal particle in approximately the following order and by the following processes at a moderate heating rate and final temperature and at atmospheric pressure. Carbon dioxide escapes under the process of decarboxylation which is the removal of the -COOH radical. The processes of decarbonylation and ring rupture releases carbon monoxide. Methane and ethane evolve from the coal through dealkylation. Distillation

and decomposition produce tar (Wen and Dutta, 1979). Tars are operationally defined as any room temperature condensible volatiles (Suuberg, 1985).

The residual char particle following the second stage is roughly spherical, more porous and may have more or less surface area than the raw particle (see Figure 1). Its mass is enriched in carbon and depleted in oxygen and hydrogen and still contains some nitrogen, sulfur, and most of the mineral matter. The particle may cool following pyrolysis causing the gaseous tars to condense or it may oxidize. In the third stage, when temperatures are above 600 °C, there is little weight loss. The major product is hydrogen, H_2, evolved from the ring rupture of aromatic C-H bonds. If taken above about 2200 °C, the residue becomes a pure carbon with the characteristics of graphite (Berkowitz, 1979).

Mass Transfer

The mechanisms, in the second stage, of volatile evolution depends on a coal's caking properties. Coals that soften, swell and resolidify are termed caking coals, while those that do not soften on heating are said to be non-caking. Generally, the higher the rate of heating, the more fluid coal becomes, and the wider the temperature range over which fluidity is observed (Berkowitz, 1979). From a table of the NCB coal classification system

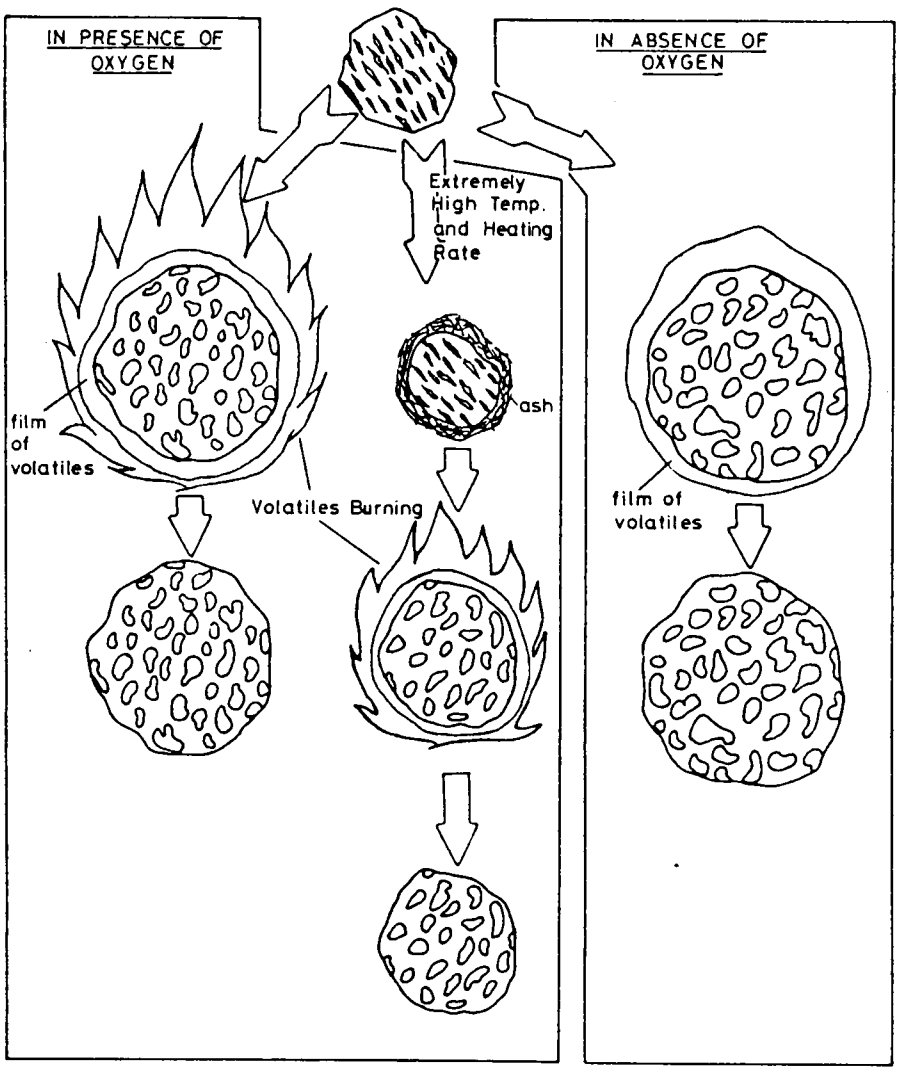

Figure 1.
Coal particles before and after pyrolysis
(Wen and Dutta, 1979)

(Berkowitz, 1979) caking properties are present in coals with volatile matter percent between 13 and 36, dry-mineral-matter-free. A broad range of bituminous coals fall in this classification. Oxidation will destroy a coal's caking properties (Tingey and Morrey, 1973). Caking coals pass through a plastic state during the second stage forming a porous char. The transport of pyrolysis products is described in terms of liquid-phase processes. In particular bubbles form, providing physical mixing as they flow outward and forming hollow spheres out of the particles because of internal ballooning. The amount of swelling is indexed by tests and is said to be rank dependent (Berkowitz, 1979).

The mode of mass transfer in non-softening or non-caking coals (anthracites, subbituminous, and lignite) generally occurs by diffusion through the pores (Hershkowitz, 1986). Pores with diameters greater than 300 angstroms and less than 10 μm are macropores; micropores have diameters smaller than 12 angstroms; and transitional pores exist between the limits of the former two. Non-caking coal particles may physically crack when heated. This is due to the internal pressure build up in the particle that occurs from rapidly forming volatiles in the solid.

Considerable pressures can build up in the particles during pyrolysis. Essenhigh (1979) suggests values as high as 1000 atm. If a coal particle does not physically crack, the gases exit forcibly and leave blowholes in the char. With small particles, the effect is to propel them rapidly and erratically through the ambient gas. If air surrounds the particle and is hot enough, the volatiles will ignite and the volatile jets will become jets of flame. As observed from experiments, by preheating the particles at about 500 °C, the jetting effect can be stopped.

Knowledge of pyrolysis has been obtained from observation and measurements in industrial practices and a great many experimental techniques. Different experiments with different pyrolysis conditions produce various results from which correlations can be derived. For example, with increasing rank, yields of tar and light oil rise progressively at the expense of water. Anthony and Howard (1976) (Figure 2) observed that there is a trend of decreasing volatile yield with increasing pressure. That trend is an example of a coal pyrolysis experimental correlation. High temperature tars are more chemically homogeneous than low temperature tars, which rarely hold any one component in amounts greater than 0.5 percent (Berkowitz, 1979). For a range of small particles,

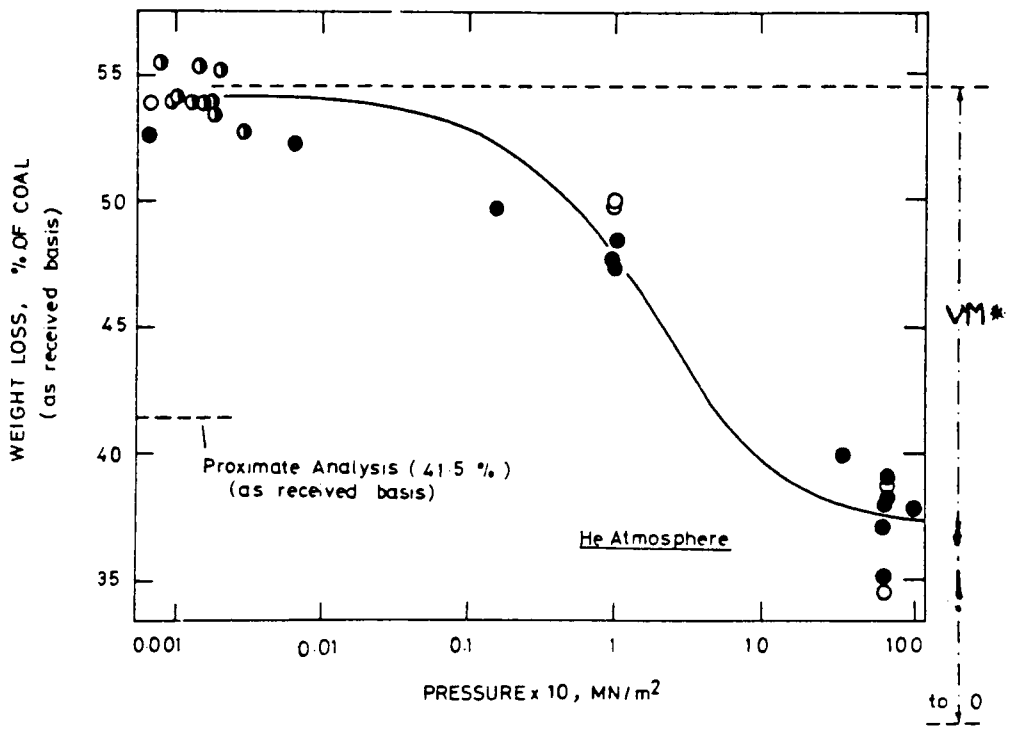

Figure 2.
An example of an experimental correlation
(Anthony and Howard, 1976)

74-400 μm, product distribution, the rate of decomposition, and the weight loss does not vary significantly with particle diameter. To provide useful results for practical use, numerous correlations have been made between the factors involved. The methods of charting relations are the results of industrial practices and a great many experimental techniques.

Experimental Methods

Pyrolysis experiments are carried out for different reasons and to obtain a variety of information. Investigators usually try to meet one or more of the following objectives:

1. to demonstrate the use of an apparatus for pyrolysis,
2. to determine weight loss,
3. to identify pyrolysis products,
4. to correlate coal temperature-time data to study kinetics,
5. to observe particle restructure,
6. to define heat and mass transfer mechanisms.

Initiative and inventiveness have produced numerous different ways to study pyrolysis. Pyrolysis experiments may be divided into two classes of methods depending on whether the particle is held captive or is carried by a

flow. Both classes support the modeling of the pyrolysis process.

Captive Sample Methods

The industrial carbonization of coal is well over 100 years old, and the aromatics derived from coal tar in Germany and England were the basis for the early synthetic organic chemicals industry. Ethylene was produced commercially from coke-oven gas as early as 1920. This and the steam power plant industry led the American Society for Testing Materials (ASTM) to standardize coal types based on their proximate analysis (volatile yield) and their free swelling index. A weighed sample of coal particles, placed in a crucible, is heated in a furnace in one atmosphere of air at 15-20 °C/sec to 950 °C. The quantity of volatiles is determined by weight loss and the resulting "button" of coal is compared to a series of standard swelling profiles.

Figure 3 shows Feldkirchner and Johnson's (1968) apparatus for a thermogravimetric analysis. A wire mesh basket was lowered into a temperature zone of 704-927°C. The coal's weight and the heating rate could be continuously measured with this device. Wiser (Wiser et al., 1967) placed a coal sample in an aluminum foil pan and admitted a stream of hot nitrogen gas to flow over it to obtain the data for a nonlinear mathematical model.

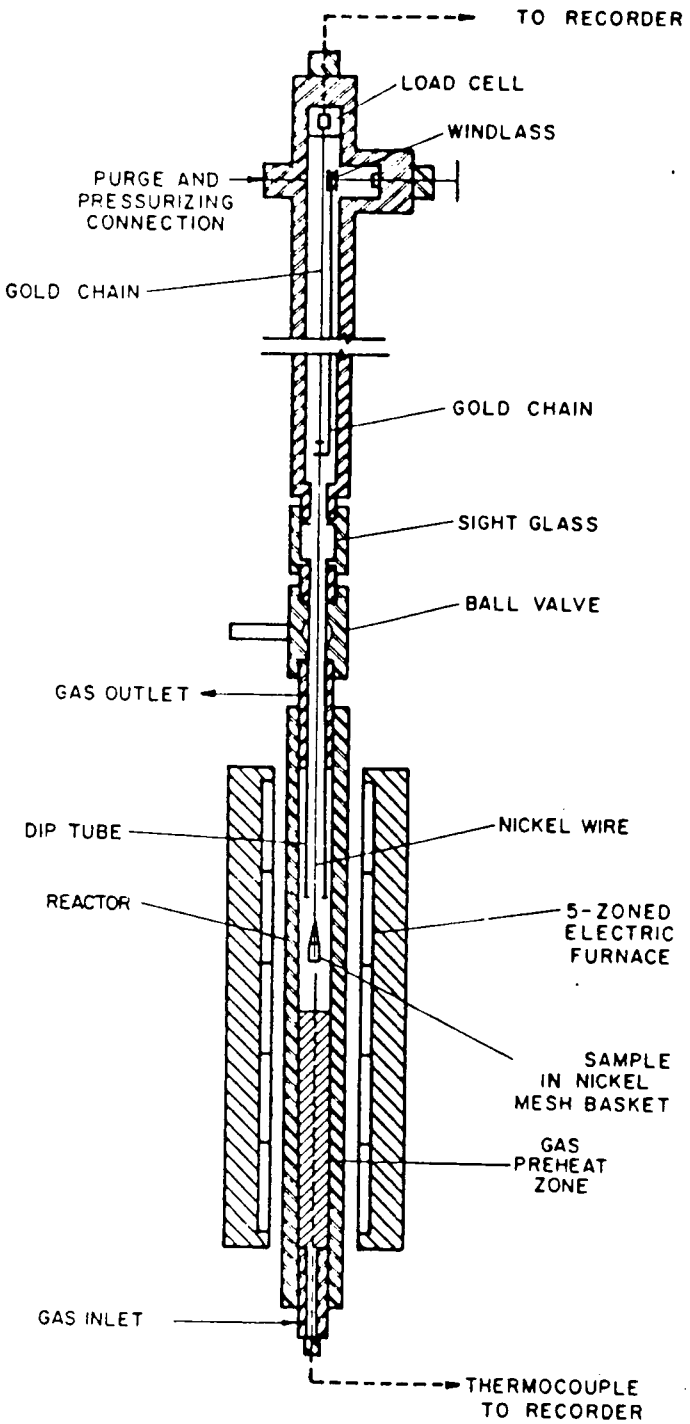

Figure 3.
The thermobalance method
(Feldkirchner and Johnson, 1968)

Sharkey and Shultz (1967) aimed broad-band and laser radiation in a pulsing manner at a coal sample sealed in a pyrex tube (see Figure 4). A crater is formed where pyrolysis takes place and the gases can be analyzed. The heating rate and temperature were not known exactly but were high in the localized area and the pressure of several ambient gases were varied, producing different gas yields.

A popular captive sample technique, sometimes called the captive sample technique, uses an electrically heated metal screen. A sample of coal particles is folded in a wire-cloth screen which is heated by a controlled electric current passing through the screen. The heating rate and final temperature can be controlled by regulating the current, though there is an uncertain heating lag time between the screen and the coal. Loison and Chauvin, pioneers of this work, in 1964 heated the screen at 1500 °C/sec to 1050 °C and assumed the sample heated rapidly along with the screen (Field et al., 1967). The assembly is encased in a shell containing an inert gas, hydrogen, or vacuum and serves as a product collection vessel (see Figure 5).

Variations and improvements have been added to the method. Some placed a sample in a electrically heated grid basket so that continuous weight measurements could

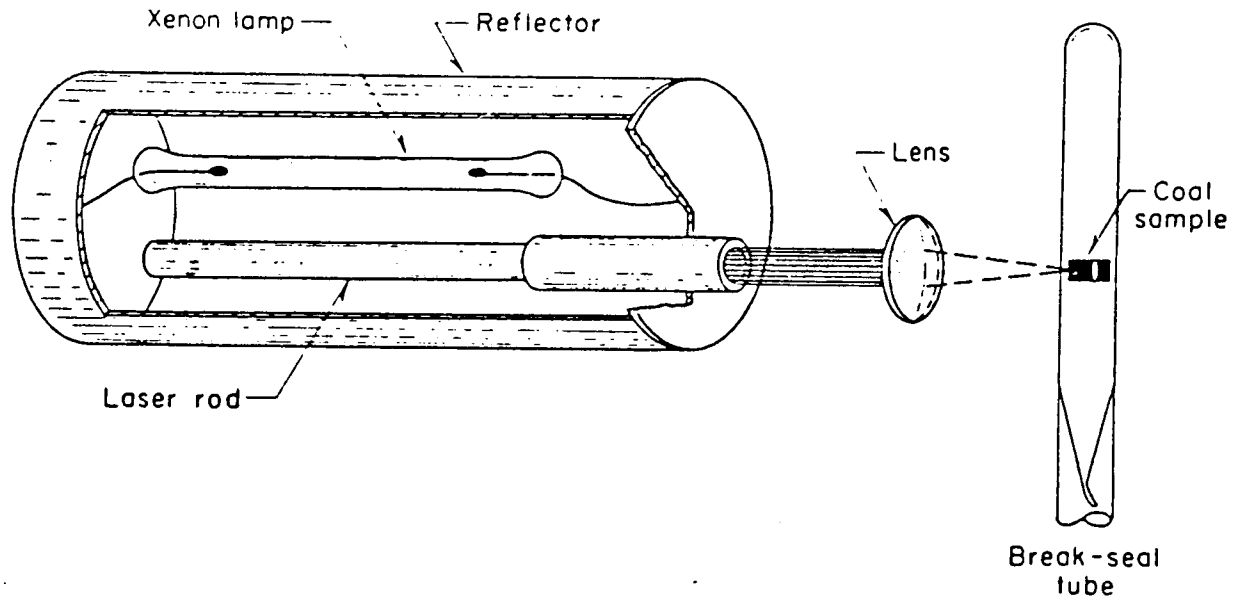

Figure 4.
Laser irradiation of coal
(Sharkey and Shultz, 1967)

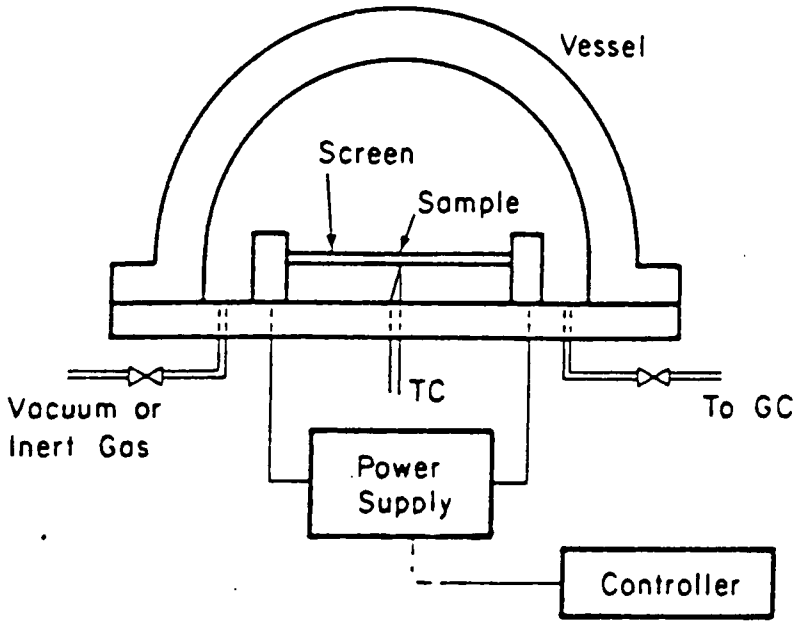

Figure 5.
The folded screen method
(Gavalas, 1982)

be done using a microbalance. Anthony et al. used a dual heating circuit to obtain heating rates of 100-12,000 °C/sec and final temperatures up to 1100 °C. The initial coal/gas ratio was small enough to ensure large dilution of the volatiles in order to retard cracking and carbon deposition on the screens (Anthony and Howard, 1976). Horton (1979) presented evidence to suggest that the screen does not catalyze secondary reactions.

The products are trapped in the vessel for collection. Suuberg et al. (1978) analyzed the volatiles with gas chromatography after lignite had been heated at 100-10,000 °C/sec to 150-1100 °C in a helium atmosphere. Coal temperatures of 1800 °C and heating rates in excess of 10,000 °C/sec for the highest temperatures can be achieved using a tungsten screen. Gas evolution kinetics are determined with Fourier Transform infrared (FT-IR) spectroscopy (Suuberg, 1985). Most of the early experiments used a metal shell, but Suuberg used a Pyrex glass vessel. The weight percentages of the products from the coal are determined by first weighing the sample, then the tar, which accumulates on the inside of the glass, and the char, which remains on the screen. The results of the methods using densely packed beds are subject to much coal particle interaction.

The pyroprobe is a platinum ribbon heating element on which a coal sample rests. As the particles are heated at up to 20,000 °C/sec to 2000 °C, a stream of inert gas sweeps the products away for gas chromatography readings (Gavalas, 1982). The pyroprobe technique assumes the loss of heavy products but provides greater speed in analyzing the gases.

There have been a few single particle captive sample studies where particles are cemented onto fine thermocouples, fine wires of different metals, and onto quartz fibers. Essenhigh (1963) conducted an experiment to determine the influence of coal rank on the burning times of single captive particles. The phenomena observed is a good illustration of the pyrolysis of coal in a reacting environment. Previously weighed particles were cemented to fine silica threads with a high temperature cement. The particles were then suspended, cantilever fashion, halfway between two horizontal heating coils so that they received radiation from both coils but convected hot air only from the lower one. The particles were placed in position before switching on the current to avoid shock heating, which tended to make the particles explode. Anson et al. (1971) suspended single particles from a quartz filament and focused radiation from two 1000 watt projector bulbs onto the particles to heat them.

A Fastax camera recorded the process at about 800 frames/sec. The objective of the study was to observe structural change. The particles were seen to soften, swell, and eject dense clouds of volatile material during pyrolysis, but as the experiment was conducted in air at 1 atm, the volatiles then burned followed by the char combustion. The creation of large blowholes in the surface was also observed on some particles. With a single particle experiment, no interaction among particles occurs. Two drawbacks to a single captive particle method are the possibility of the thread acting as a heat sink or catalyst and particle size is restricted to relatively large sizes.

Coal Flow Methods

Flow techniques continuously feed and withdraw coal from the experimental apparatus. Fluidized bed and entrained flow studies have close applications to industrial gasifiers. These experiments usually are based on large size equipment by scaling down the reactors to laboratory sizes. Flow methods have the potential for high heating rates and accurate volatiles analysis. However, photography is inconvenient because the particles do not exist in a given plane of focus.

After fluidized beds had been used for coal pyrolysis for a number of years, Jones et al. (1964) ran a reactor

for a variety of conditions to find optimum conditions. His apparatus was the forerunner of the Char Oil Energy Development (COED) process. A schematic drawing of the fluidized bed reactor system is shown in Figure 6. Coal is preheated and suspended in a vertically rising current of fluidizing gas. The bed is stationary but the upward flow of air and gas causes extreme turbulence resembling a boiling liquid. The technique provides rapid heat transfer to the particles and the final temperature can be over 1500°F. Product analysis was easy for char, (recovered in the solids receiver), and for gas, (piped through an analyzer and metered), but the tar was found throughout the system. The system had to be weighed in sections to determine the net tar yield.

Difficulties arise in this technique because mineral matter builds up in the bed interfering with the decomposition rate. Bituminous or swelling coals, injected into a hot bed, become plastic and, in contact with similar particles, agglomerate to form larger particles. The bed may then collapse and quench the material into hard deposits. Preoxidation helps the coal by reducing caking properties. Kayihan and Reklaitis (1980) describe a design model for a coal pyrolysis system involving sequential bed reactors with recycle char streams to provide fluidized gas for upstream beds.

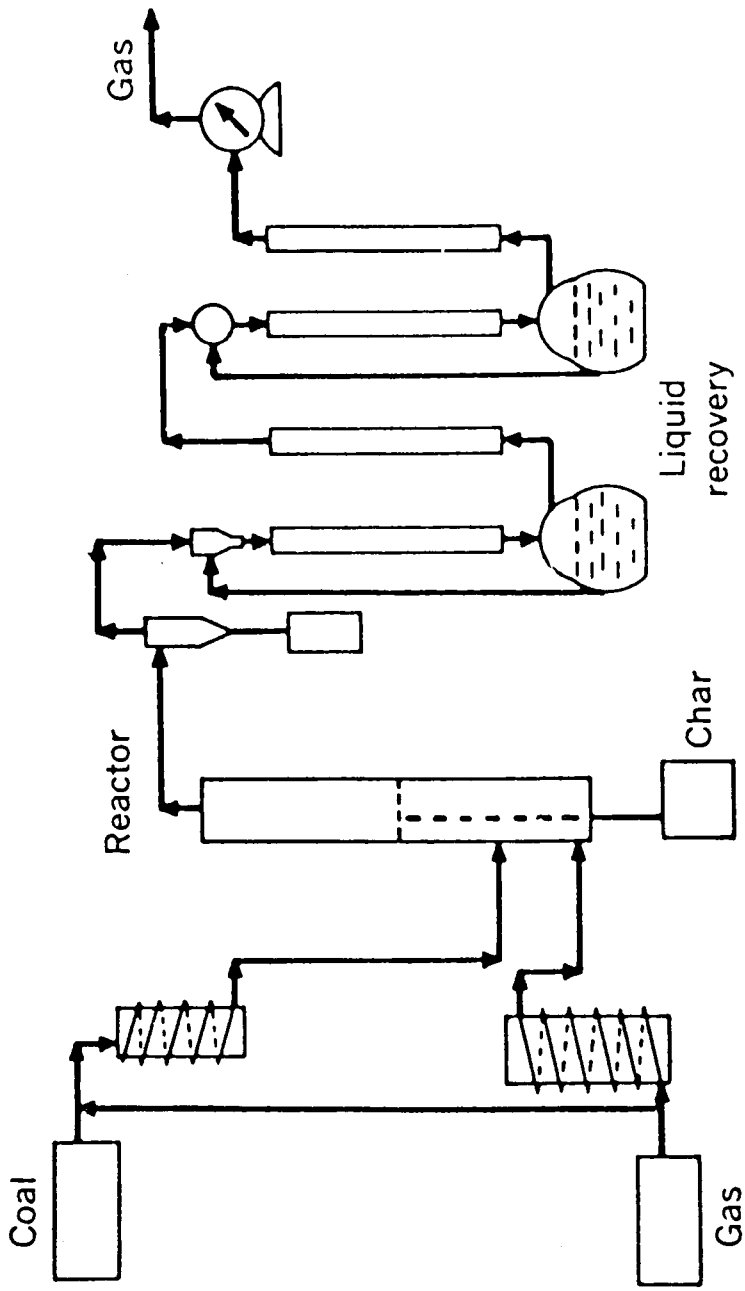

Figure 6.
The fluidized bed method
(Jones et al., 1964)

In an entrained flow experiment, coal particles, entrained in a stream of inert gas, flow vertically through a furnace. From a parallel input, secondary inert gas is injected into the stream at a Reynold's number low enough to assure laminar flow and to act as a buffer; the particles will not disperse radially to the walls, (the particles' velocity is axial). Advantages of this setup are the collection of products, the high heating rates attained, and the reduction of agglomeration tendencies found in fluidized beds.

Kobayashi et al. (1976) used a stream of argon gas and graphite heating elements to achieve heating rates of 10^4-$2x10^5$ K/sec and temperatures of 1000-2100 K. It was shown again that pyrolysis yields a greater weight loss in less time at higher heating rates and higher final temperatures than the ASTM proximate analysis yield. Nsakala et al. (1977) heated particles in an entrainment of nitrogen. They were looking for data to support the mathematical pyrolysis "Two Component" model. The design of the reactor dated back to 1979 and yielded rates of 8000 °C/sec. Chang et al. (1980) built a scaled down gasification reactor in the Garrett Flash Pyrolysis process mold (McMath et al., 1974). Pulverized coal was fed into a mixing chamber where it contacted hot recycle char and immediately moved into the entrained flow reactor

where it flash pyrolyzed within 2 sec at ~870 °C for gas production or at ~570 °C for oil.

The entrained flow technique provides for methods of product collection. Because caking coal particles are prohibited from sticking to the walls by their axial velocity, the coal that enters, exits in its components. The char, tar and gases are separated after quenching by cyclones and filters. Gas chromatography is used to analyze certain of the gases. FT-IR spectroscopy can characterize the char. Solomon et al. (1986) (Figure 7) also directly measured particle temperature and velocity by using FT-IR transmission spectroscopy. Since coal particle temperatures had generally not been directly measured during rapid pyrolysis, more accurate kinetic data were obtained.

In order to eliminate the calculations involved with stream velocities and to solve some of the problems that result from using a captive sample, particles can undergo pyrolysis in free fall. With the use of a drop tube, coal is injected at the top and is heated by radiation as it falls at terminal velocity downward. Studies have incorporated heated furnace tubes, broad band lamps, lasers and various combinations for particle heating. Since the particle is radiatively heated, it need not contact a solid surface, be it the container or another

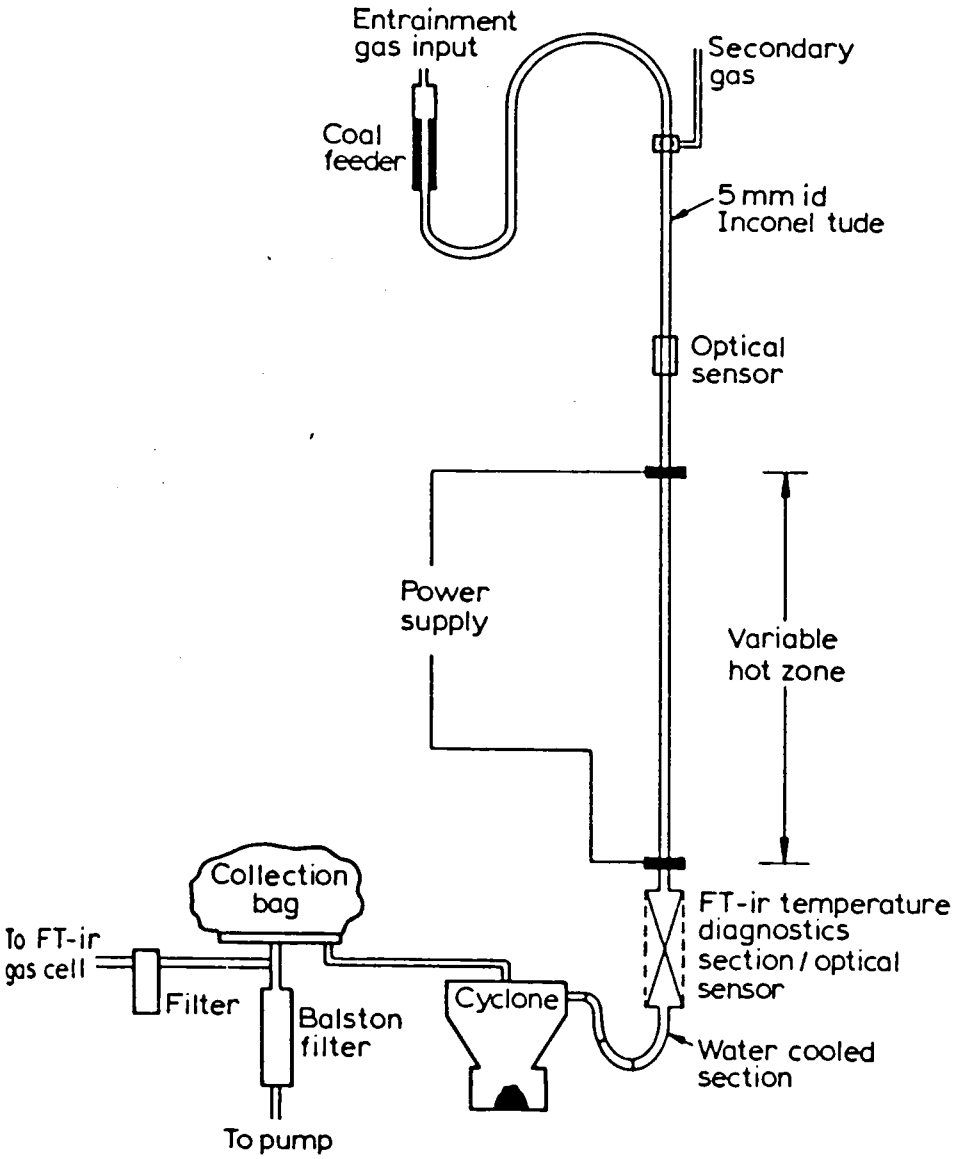

Figure 7.
The entrained flow method
(Solomon et al., 1986)

particle. Though limited by the materials of the flow tube, high heating rates are possible. Gat et al. (1983) observed rates between 10^5 and 10^6 K/sec and final temperatures between 2000 and 2900 K in a laser heated environment using a three color pyrometer. Products may be analyzed at the bottom but damage to the char particles caused by their collection and the inability to observe the pyrolysis process in free fall are the drawbacks to the drop tube method.

A method of observing the behavior of coal particles during thermal decomposition is given by Samuelson et al. (1981). Particles carried by air were injected into a flat methane-air flame. Using a holographic system, the coal could be imaged in a dynamic field. Particle restructuring was observed along with clouds of volatiles as pyrolysis took place, but it was not possible to identify a particle's history throughout the process.

A survey of pyrolysis experiments has been made. Captive sample and coal flow techniques, their operating methods, drawbacks and some of the results have been discussed. The drop tube method seems to be the superior process because it provides a wide range of heating rates and accurate product and temperature analysis. The results from the experiments are used to support the

mathematical models derived to describe the behavior of coal during pyrolysis.

Pyrolysis Modeling

A correlation of experimental data for mass loss by pyrolysis is shown in Figure 8. This figure shows that coal first loses weight rapidly upon heating, then the weight loss rate decreases. The constant temperature curves show that increased weight loss with increased temperature. Anthony and Howard (1976) describe simple reaction models used by many authors approximating these curves as a first order decomposition with the rate described by

$$dVM/dt = k(VM^* - VM). \qquad [2]$$

VM* is the effective volatile content of the coal, usually the proximate analysis volatile yield, so VM* varies with coal type and is determined empirically. The rate constant k is typically the temperature dependent Arrhenius expression

$$k = k_0 \exp(-E/RT), \qquad [3]$$

where the activation energy can be taken as roughly 32,000 cal/mol (Fuchs and Sandhoff, 1942). The first order model has been contested as being inadequate. Wiser et al. (1967) found that a second order reaction equation fit their data more closely:

$$dVM/dt = k'(VM^* - VM)^2 . \qquad [4]$$

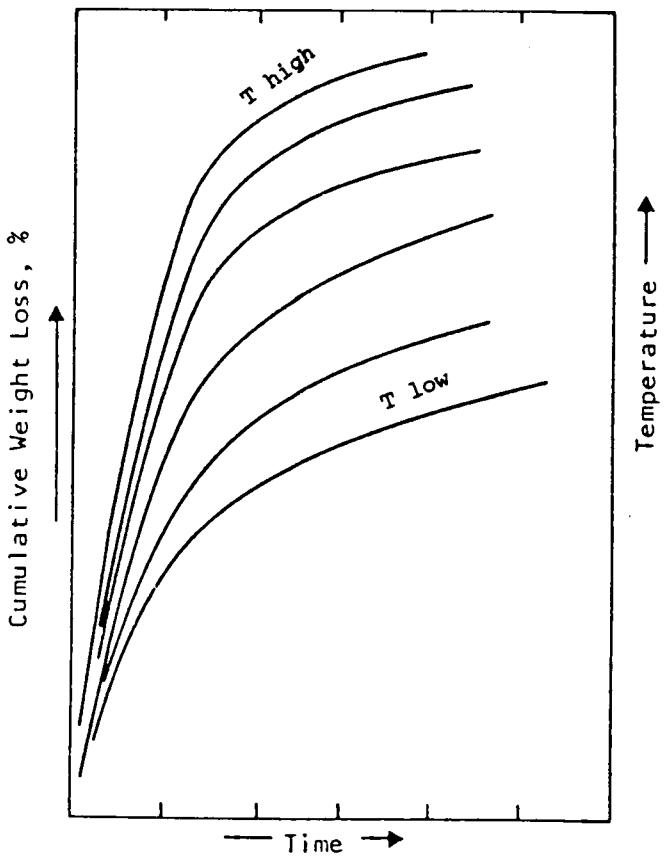

Figure 8.
Isothermal weight loss vs. time curves
(Berkowitz, 1979)

However, neither scheme fits the empirical data at the higher temperatures.

Kobayashi et al. (1976) suggested that pyrolysis could be modeled with a pair of first order irreversible reactions with different activation energies dependent on the specific coal dust. Others, expanding on the idea, assumed the thermal decomposition of a coal particle consisted of a large number of independent parallel rate reactions. Anthony used an infinite series of reactions assuming a continuous Gaussian distribution of activation energies. This model correlated well with Anthony's experimental data and with Suuberg's data (1978) which was determined using the folded screen technique.

For a full prediction of pyrolysis, a model should produce the composition of the volatile gaseous products and the char, in addition to the rate of volatiles evolution. The Suuberg et al. (1978) model provides for specific rates for some compounds but cannot characterize some tars and hydrocarbons. An assumption is made that one, two or three parallel reactions, depending on the observed behavior, describe the formation of certain key products. Kinetic parameters, including activation energies, for various products were calculated from experimental data (folded screen method). Product

rate formation curves were calculated using these parameters and the parallel reaction model.

The purpose, required accuracy and viewpoint of the modeler of coal decomposition influence the choice of a coal pyrolysis model. The simple reaction models assume that coal is sufficiently homogeneous that it can be treated as a material which degrades thermally in one step into volatile matter and carbon residue. This approach is useful where pyrolysis rates do not dominate the process and where appropriate data can be incorporated for the exact condition being modeled. At the other extreme, if coal is assumed highly heterogeneous, pyrolysis has to be treated essentially as a statistical assembly of reactions because the various constituents decompose independently, then are broken down further. This treatment is used if composition and rates are critical to the process. An intermediate course, useful for some engineering purposes, is to use a modification of the two reaction model. George and Laurendeau (1982) coupled atom balances and typical values of methane production with the Kobayashi model. CO, CH_4, C_2H_2, C_2H_6, H_2O, NO and SO_2 were their predicted pyrolysis products and increasing temperature increased the aromatic content of the products (George, 1984).

Most of the models presented here and elsewhere are based on the results of the electric grid or entrained flow experimental techniques. The drawbacks discussed relating to these methods might carry over to the models. Further investigations with the use of an acoustic levitator could extend the available data to include higher temperatures and heating rates.

Acoustic Levitation

The second topic of this literature review is acoustic levitation because this research incorporates its use to study coal pyrolysis. Acoustic levitation, as it relates here, is the suspension of solid or liquid matter in a gaseous atmosphere using sound waves. Perhaps the earliest record of acoustic levitation is in 1940. Hillary St. Clair was "astonished" when a penny or lead shot could be suspended in midair, supported only by sound radiation pressure (St. Clair, 1941). He was able to deliver 200 watts of electrical power to a vibrator operated at 10-20 kHz.

In the operation of a single sound source levitator, a sound field must be produced that will suspend solid or liquid matter in a gaseous medium. To accomplish this, a driving piston radiates a beam of sound toward a parallel reflecting surface. The reflector is placed a distance $n\lambda/2$ away from the driver, where λ is the sound wavelength

in the medium and n is any integer (see Figure 9). With an arrangement of this type, a standing wave is produced with surfaces of minimum pressure, called nodes and also referred to as "acoustic wells." Lee and Feng (1982) may have used the word "well" to describe a surface because when a liquid or solid particle is introduced near one of the surfaces, it will "fall" into a suspended position.

The particle's levitated position in the sound field is wherever the acoustic potential energy is a minimum. The phenomenom which maintains a particle's position can be described as a potential energy, a pressure, or a pressure gradient force. In terms of forces measured on a levitated body, the pressure gradient force in the vertical direction overcomes gravity. Horizontal pressure gradient forces position a body laterally. Since gravity will cause a particle to be below the node (there is no net acoustic force on a precisely centered, symmetric particle), the body's height must be significantly smaller than $\lambda/2$ so that it does not overlap successive levitation nodes.

The greatest levitation ability occurs when the driver and the reflector are operated at their resonant frequency (Oran et al. 1980). The minimum sound pressure level (SPL), which can be expressed as the root mean

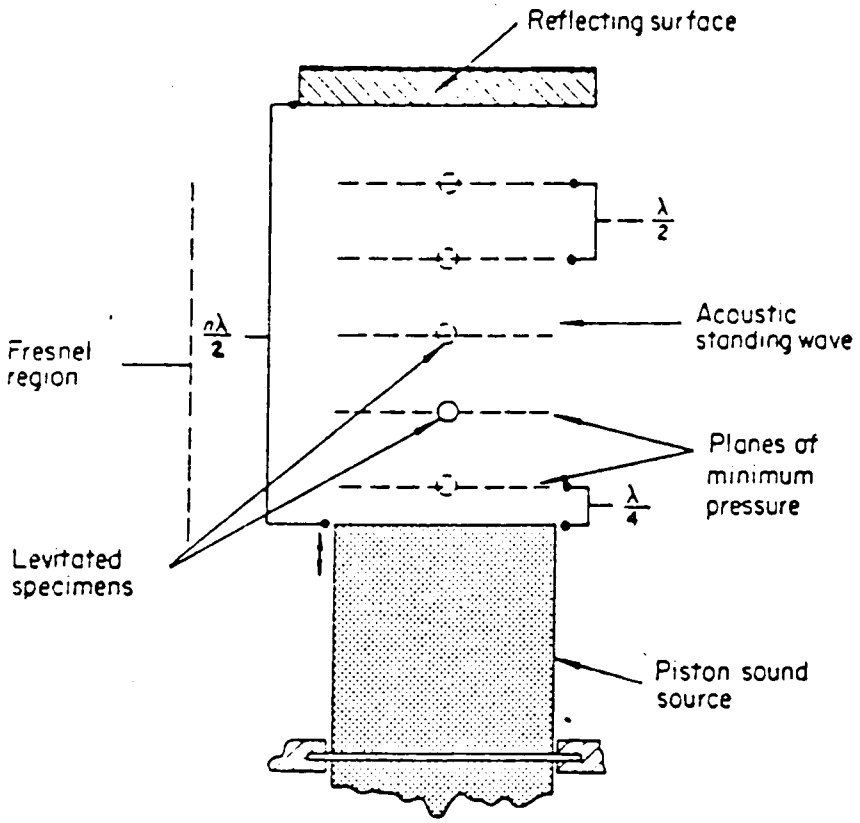

Figure 9.
An energy well levitator
(Whymark, 1975)

square pressure, required to levitate a small particle is

$$P^2_{rms} = (2\lambda c^2 \rho \rho_s g)/5\pi \quad , \qquad [5]$$

where c is the speed of sound in the gaseous medium, ρ and ρ_s the medium and particle densities respectively and g gravitational acceleration (Lee and Feng, 1982). The minimum SPLs required range at 50 kHz from 154 dB for wax, 156 dB for coal, to 168 dB (Re $2x10^{-5}$ N/m^2) for gold.

Varying the geometry of the reflector or driver can change the wave. By machining a concave on the reflecting surface, the pressure surfaces are curved producing increased lateral forces on the particle but decreased vertical forces. The focused sound energy is analogous to focused light in geometrical optics. The smaller the radius of curvature of the concave, the greater the focusing effect until the radius of curvature equals the radius of the reflector.

Acoustic streaming exerts an additional force on the sample. The viscous drag force of streaming is caused by vortices or time-dependent circulation in the medium. It depends on the geometry of the driving arrangements, schemes for the standing wave production, and other unavoidable acoustic boundaries (Lee and Feng, 1982). To prevent the levitated sample from being ejected from its stable position, the streaming force must be small compared to the fundamental volumetric force.

The efforts made in developing an acoustic levitation apparatus have been mainly aimed toward use in space. The fields and forces necessary to counteract accelerations of 10^{-4} g on a spacecraft are relatively small. At the Jet Propulsion Laboratory, a system was developed for applications in space, using crossed sound beams on three axes for positioning (Barmatz and Jacobi, 1983). Figure 10 is a diagram showing a triple axis system.

Earth based containerless processing can improve our readiness for the future. Heat sources, including an induction furnace, a conventional furnace, a laser beam, or a projector lamp can melt or burn materials without contamination from containers. Whymark operated a single axis levitator at 20 kHz with an electrical power input of 85 watts. A muffle furnace, capable of reaching temperatures to 1000 °C, heated various materials. The difficulties of processing on earth at high temperatures can be appreciated by realizing that the levitation force decreases by $\sim(1/T)^{5/2}$ due to a decrease in air density and an increase in the wavelength (Oran et al., 1980). The melting of aluminum and glass is described (Whymark, 1975). When a material is in its liquid state, its shape conforms to the geometry of the acoustic well. Figure 11 shows a particle of liquid wax with a spherical shape because of its surface tension and the influence of curved

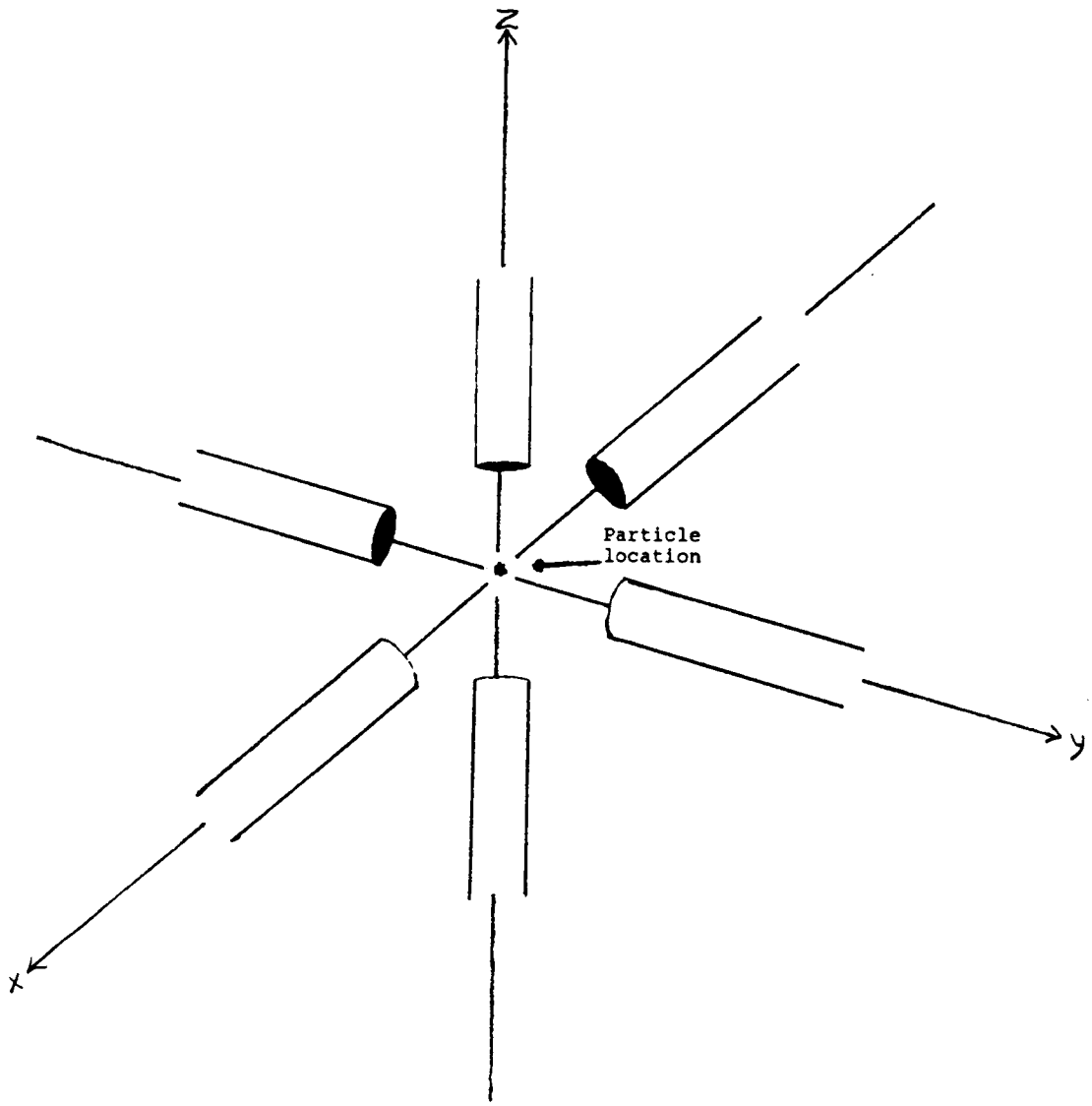

Figure 10.
A triple axis levitator

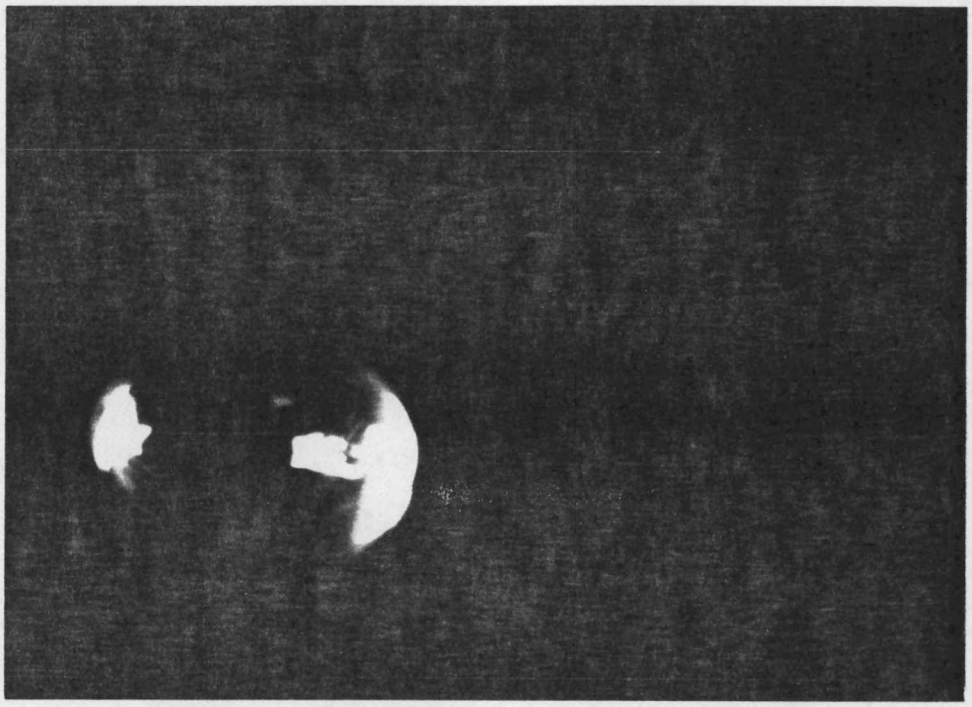

Figure 11.
A droplet of liquid wax

pressure forces. The extent of conformity depends on the material's surface tension, where samples with high surface tension resist conformity or deformation. With flat and parallel driver and reflector geometries, a liquid tends to become disc shaped. If a material's viscosity is too low, it will usually rupture and be ejected from the field (Oran et al., 1980).

Lee and Feng (1982) designed a hemispherical focusing dish driver to levitate submillimeter samples. The apparatus would resonate at 75, 107, and 163 kHz. By setting the frequency at 107 kHz and increasing the SPL to 172 dB, solid gold could be levitated. With a spherical concave reflector approximately $2\lambda/3$ in size, the lateral positional wandering of the sample in the well was less than 5 percent of the dimension of the sample size used. When an irregularly shaped piece of wax was heated and cooled, it became a sphere.

Acoustic frequencies may be audible or ultrasonic (Wang et al., 1974). Oran et al. (1980) had a device with a resonant frequency of 15 kHz to conduct a parametric study. Because sound pressure levels exceeding 130 dB are needed, operation should be in the ultrasonic region, greater than 20 kHz for most people. Audible sound of this pressure level will cause pain and rapid hearing damage.

Based on the literature reviewed, it was certain that coal particles could be levitated. Given the proper materials and electrical equipment, a levitator could be built by applying the basic principles and adjusting driver and reflector shapes by trial and error to achieve the objectives of this research. Since engineers have been able to construct levitating systems and use them for high temperature containerless processing, a containerless coal pyrolysis experiment may be demonstrated.

CHAPTER III

EXPERIMENTAL METHOD AND APPARATUS DEVELOPMENT

Chapter III describes the design, construction, and preliminary testing of the acoustic levitation apparatus and the supporting optical equipment used for this research. The levitator was designed to meet the main objective, that is to observe and photograph a coal particle undergoing pyrolysis. The reason for capturing and heating a coal particle in this manner is to avoid some of the drawbacks of previous experimental methods. A single particle can be suspended without contacting other solid bodies. Heating and lighting optics involving broad band illumination and lasers can simply be focused on a small area barely larger than the particle diameter. The viewing field for the microscope and attached camera is therefore small and stationary.

Acoustic Levitator

A review of the literature concerning acoustic levitation in air revealed three existing apparatus configurations. The triple axis (Barmatz and Jacobi, 1983) system was considered too complex and expensive. Lee and Feng's (1982) focusing dish, although specially designed for submillimeter samples, appeared to be more complex than necessary. A single axis driver-reflector

configuration was chosen (Whymark, 1975) for its simplicity. Particles having densities near that of coal (1.2 g/cc < density < 2 g/cc) had been levitated. A single axis system allowed simple optical access for monitoring and heating.

Before discussing the specific design of the levitator it is necessary to discuss the nature of piezoelectrics because of their importance in the project. Piezoelectric transducer (PZT) discs, manufactured by the EDO Corporation, are made of a lead titanate zirconate composition of ceramic. They are polarized by aligning their crystals parallel to an electric field giving the ceramic specific electrical and physical properties. They are silvered on the top and bottom to provide for electrical connections. Piezoelectrics have the ability to convert electrical pulses to mechanical oscillations and vice versa. They must be operated in a certain designed temperature range because of thermal expansion and depolarization.

A prototype levitator was built using materials and electrical equipment available at the University of Tennessee (see Figure 12). A Wavetek signal generator, with a 0-1 MHz capacity, fed an approximately 24 kHz oscillation to an amplifier (a military surplus public address audio amplifier was used). The voltage was then

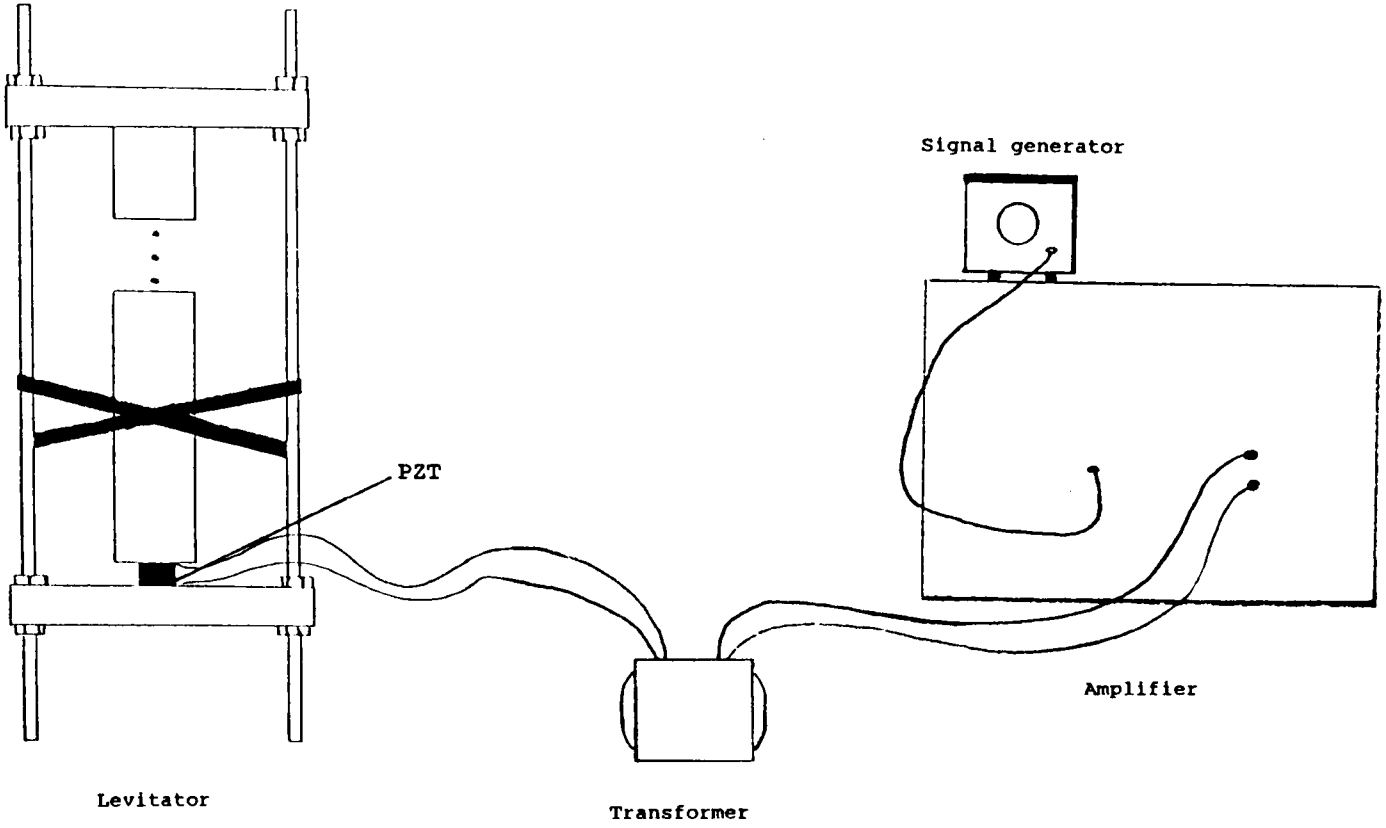

Figure 12.
The prototype levitator

stepped up via a transformer and the two electrical leads soldered to opposite sides of a piezoelectric transducer (PZT) disc (approximately .5 in x .75 in diameter).

The PZT disc was an element of the sound generator or driver. It was attached with cyanoacrylate (Super Glue) to the bottom of an aluminum cylinder (approximately 10 in x 1.5 in diameter). Supported at its midpoint by strong rubber bands, the cylinder and attached PZT disc were forced against a plexiglass base at the bottom. When the voltage was introduced, the cylinder transmitted sound waves out the free end. By glueing a smaller aluminum cylinder (3 in x 1.25 in diameter) to a plexiglass support panel in a parallel formation a small distance (14.3 mm) away from the driver's free end, a standing wave was produced. Because of the driver-reflector spacing, there were four nodes in the standing wave or four acoustic wells (see Figure 13). The restoring forces in each node decrease as the number of nodes increase. However, to physically place a sample into a well and to provide spacing for heat and illumination, a minimum of three wells was essential. Samples of styrofoam, with characteristic lengths of 1 to 3 mm, introduced into the field, were levitated (see Figure 14).

The initial apparatus demonstrated that the principle of standing wave generation for acoustic levitation is

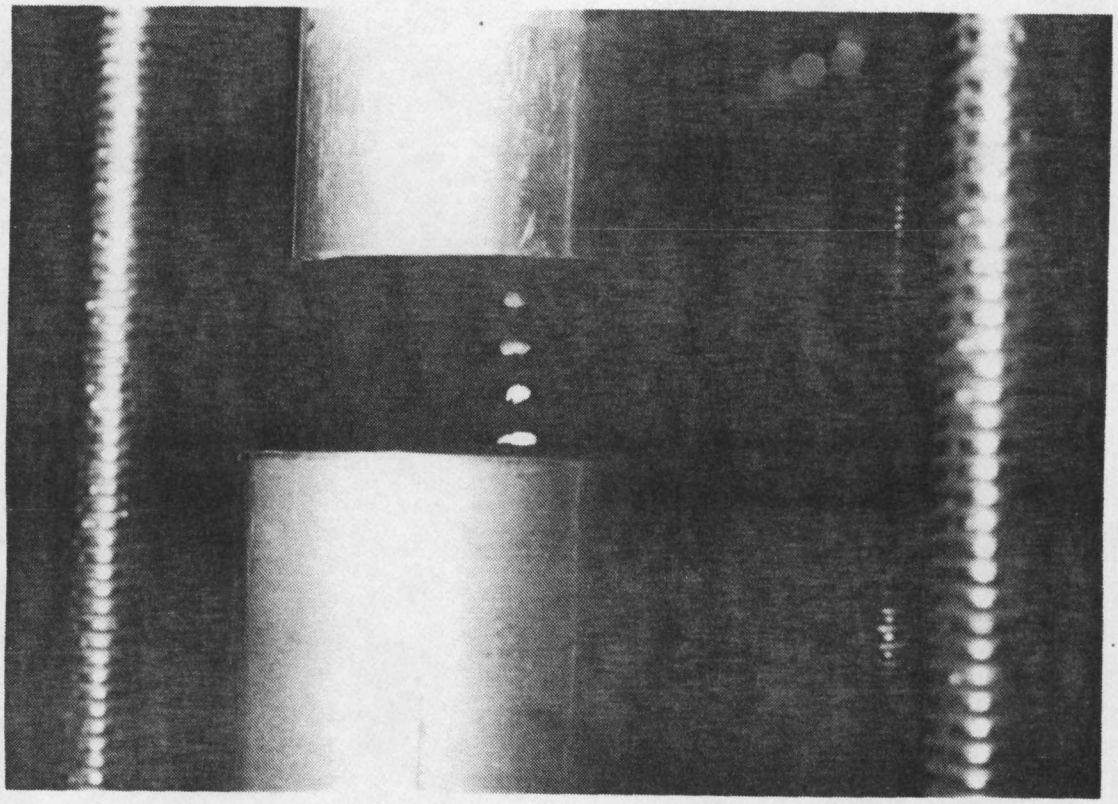

Figure 13.
Four acoustic wells in the prototype

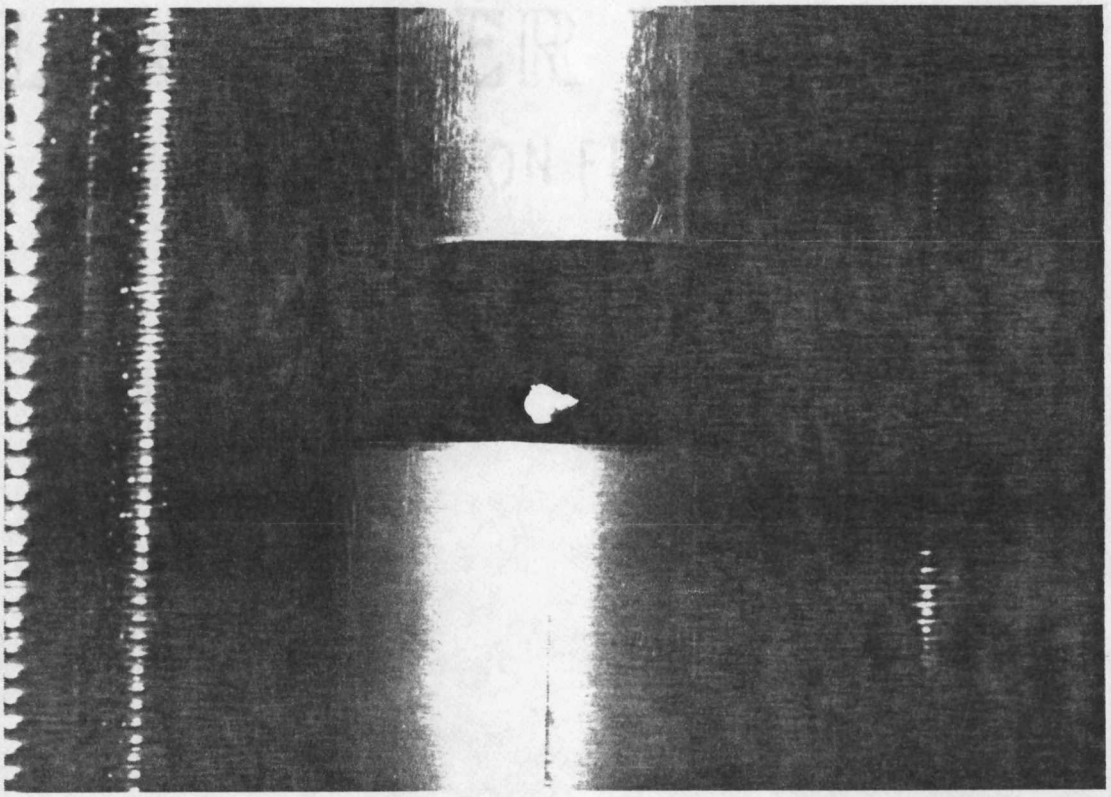

Figure 14.
A 3 mm styrofoam sample in the prototype

valid. The system was designed to send an ultrasonic wave between two flat parallel surfaces. The pressure gradient forces were adequate to levitate styrofoam but were generally too weak to suspend coal. On occasion, materials other than styrofoam could be levitated, but at alternate times, undesired noisy vibrations would occur as the driver rattled against the base.

Refinement of the apparatus occurred through logic and trial and error. A larger PZT disc (.5 in x 1.5 in diameter) was used on the driver to give an overall greater displacement. The driver assembly was glued to the base, discarding the rubber bands, to prevent unwanted screeching from secondary vibrations. Later the driver was glued to a large bolt that could be fastened tightly to a steel base (which replaced the plexiglass base) so drivers could be removed or exchanged. Different driver geometries and sizes were designed, machined, and tried. The final driver geometry was a truncated cone (1.5 in bottom diameter, .75 in top diameter) theorized to channel the sound energy. Since the truncated cone performed better than the cylinders with flat or concave ends, it might be concluded that the sound energy from the PZT disk, after transmission up the cone, was concentrated into a smaller area.

The original reflector had been glued to a plexiglass support panel. To adjust the reflector to an $n\lambda/2$ position, there was the undesirable task of turning eight nuts. This system was replaced by a threaded rod as shown in Figure 15. By rotating the rod, the reflector could be raised or lowered, staying parallel, relative to the driver. A spherical concave was machined into the end of the cylinder reflector to increase the lateral positioning forces to counteract the effects of microjets formed in the particle by escaping gases. The radius of curvature of the concave was 1 in. Figure 15 is a sketch and Figure 16 is a photograph of the acoustic levitator used in this research.

When the equipment still did not consistently levitate coal, an investigation into the electronics revealed that the amplifier was inadequate. The voltage output dropped considerably over 20 kHz and was reduced to close to zero by 30 kHz because the amplifier is an audio unit for loudspeakers. A Krohn-Hite 75 watt wideband amplifier was rented. A Hewlett-Packard oscillation generator was used because its vernier-type dial allowed for better frequency resolution. When the levitating apparatus was operated at resonance around 41 kHz, coal could be levitated.

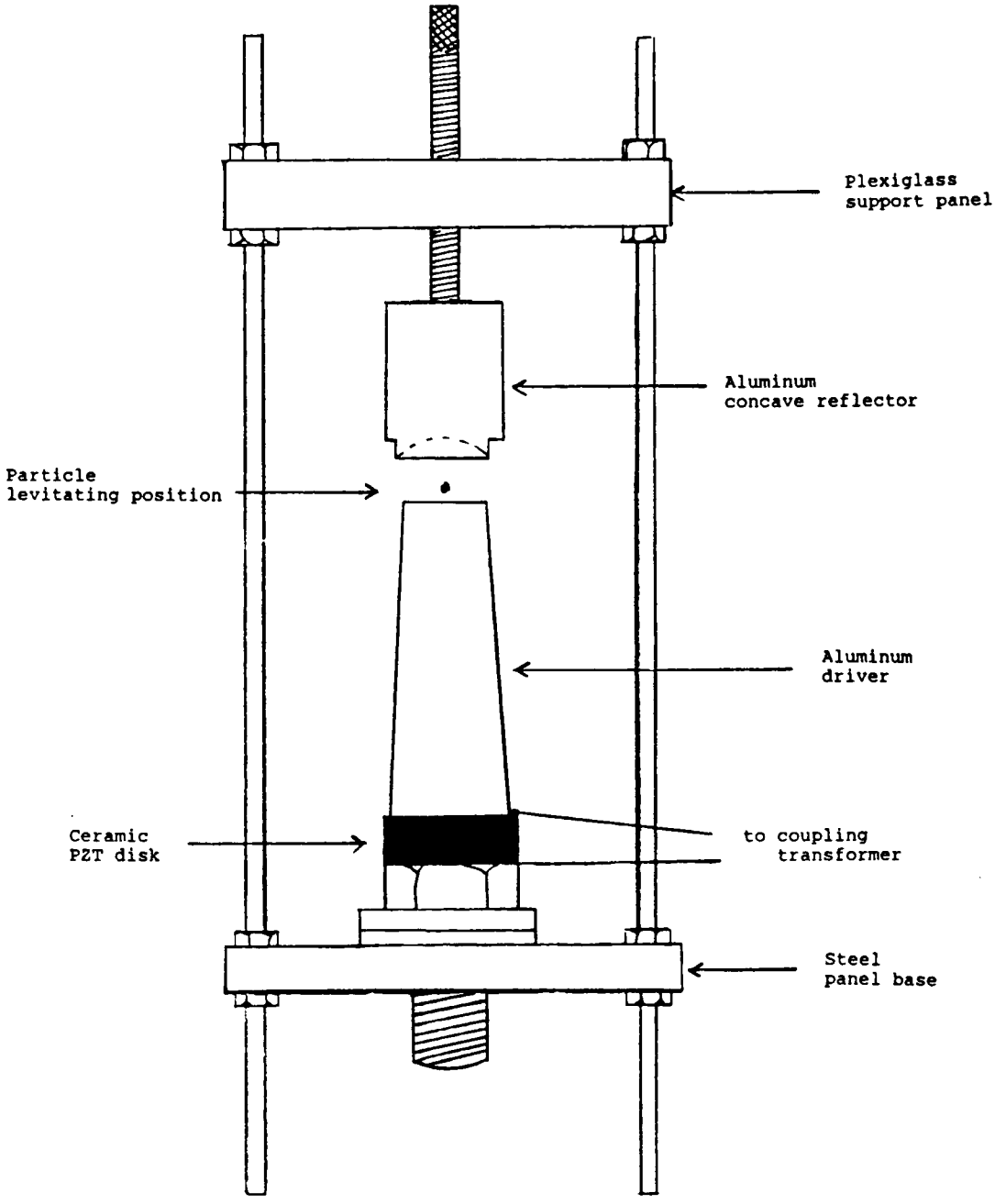

Figure 15.
The acoustic levitator

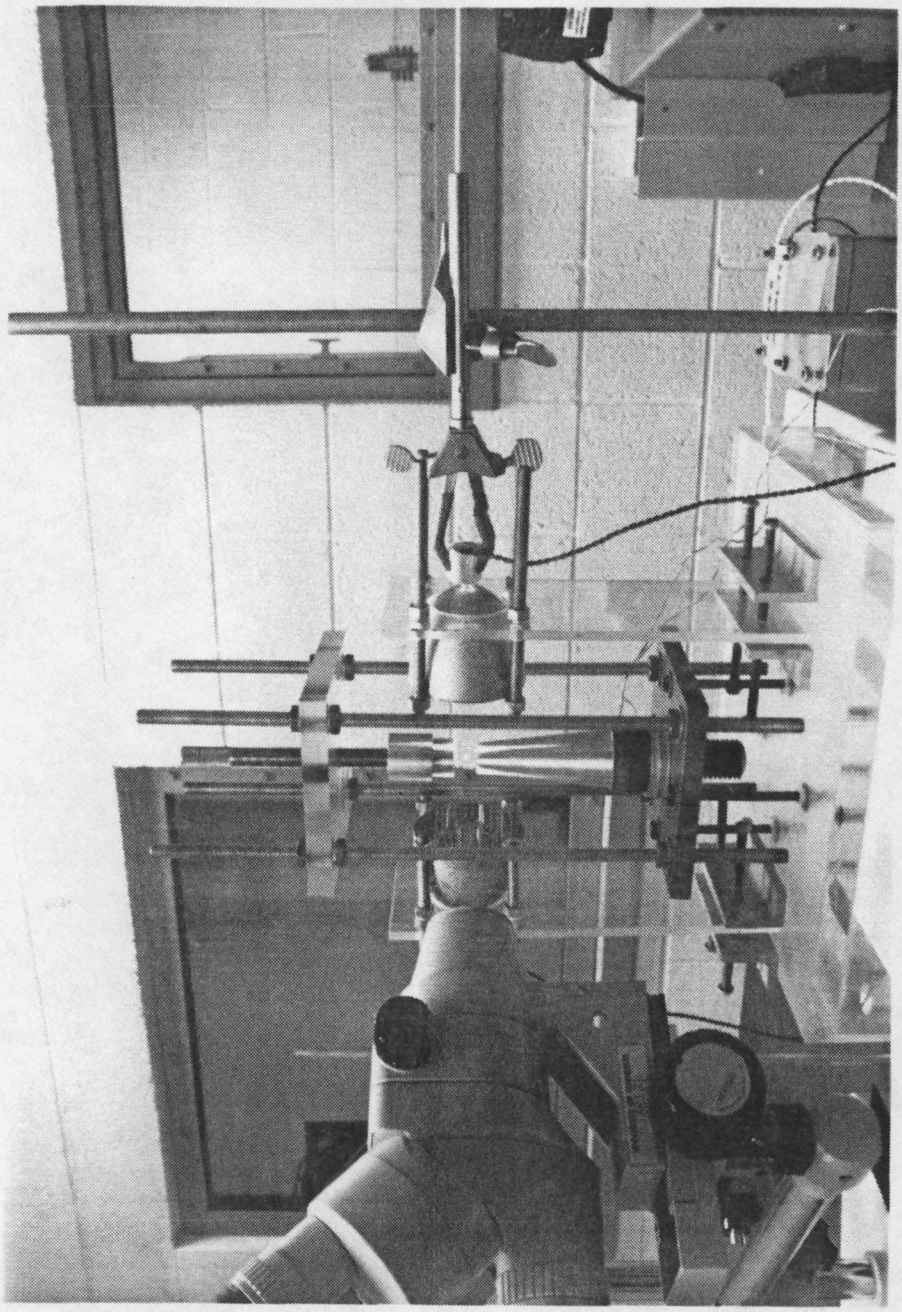

Figure 16.
A photograph of the acoustic levitator

The minimum SPL required to levitate coal at 41 kHz is 157 dB using equation [5] (page 35) with ρ_s = 1300 kg/m^3, λ = 8.46 mm and using air at room temperature. An attempt was made to measure the SPL achieved with the levitator. Readings taken from a SPL meter showed that the SPL was in excess of 130 dB because the meter's indicator needle exceeded by the scale's limit of 130 dB.

An Electronic Navigation Industries 400 watt wideband amplifier and matching transformer were purchased to replace the rented one. A power meter on the amplifier actually helped find the resonant frequency of the levitator. When the power reflected back to the amplifier was minimized, the levitator was at resonance. This amplifier has a safety feature of shutting off when 70 watts of power are reflected back to it from the levitator to prevent overload. Figure 17 is a diagram of and Figure 18 is photograph of the electrical equipment.

Optical Equipment

Figure 19 shows how the particle position in the single-axis levitator is in the center of the surrounding optical equipment. Two Aerotech 5 milliwatt helium-neon lasers are easily positioned and aimed at the particle. They provide a small portion of the energy to heat the coal particle. Two broad band GTE 12 volt, 50 watt tungsten-halogen lamps provide most of the heating

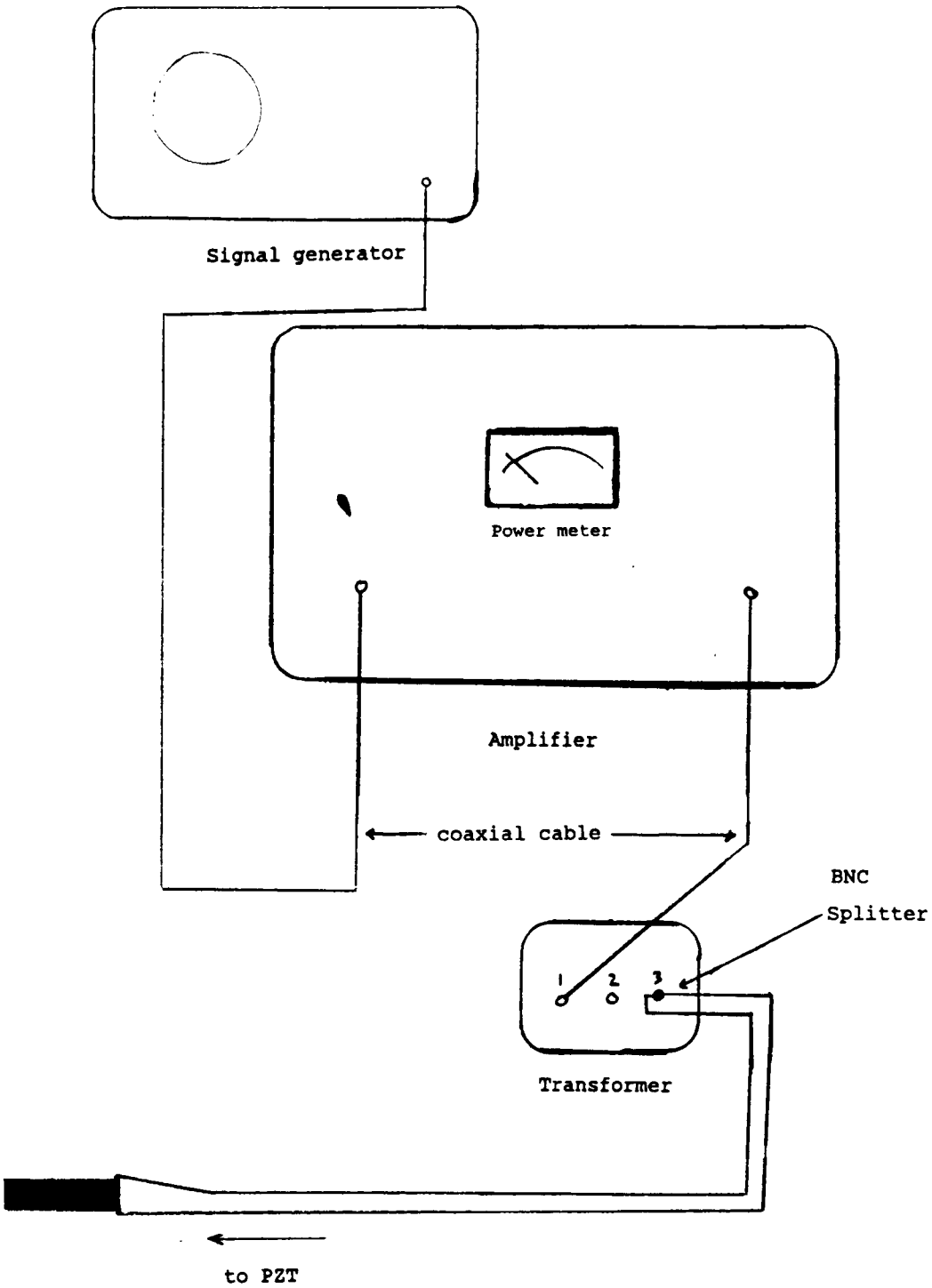

Figure 17.
The levitator electronics

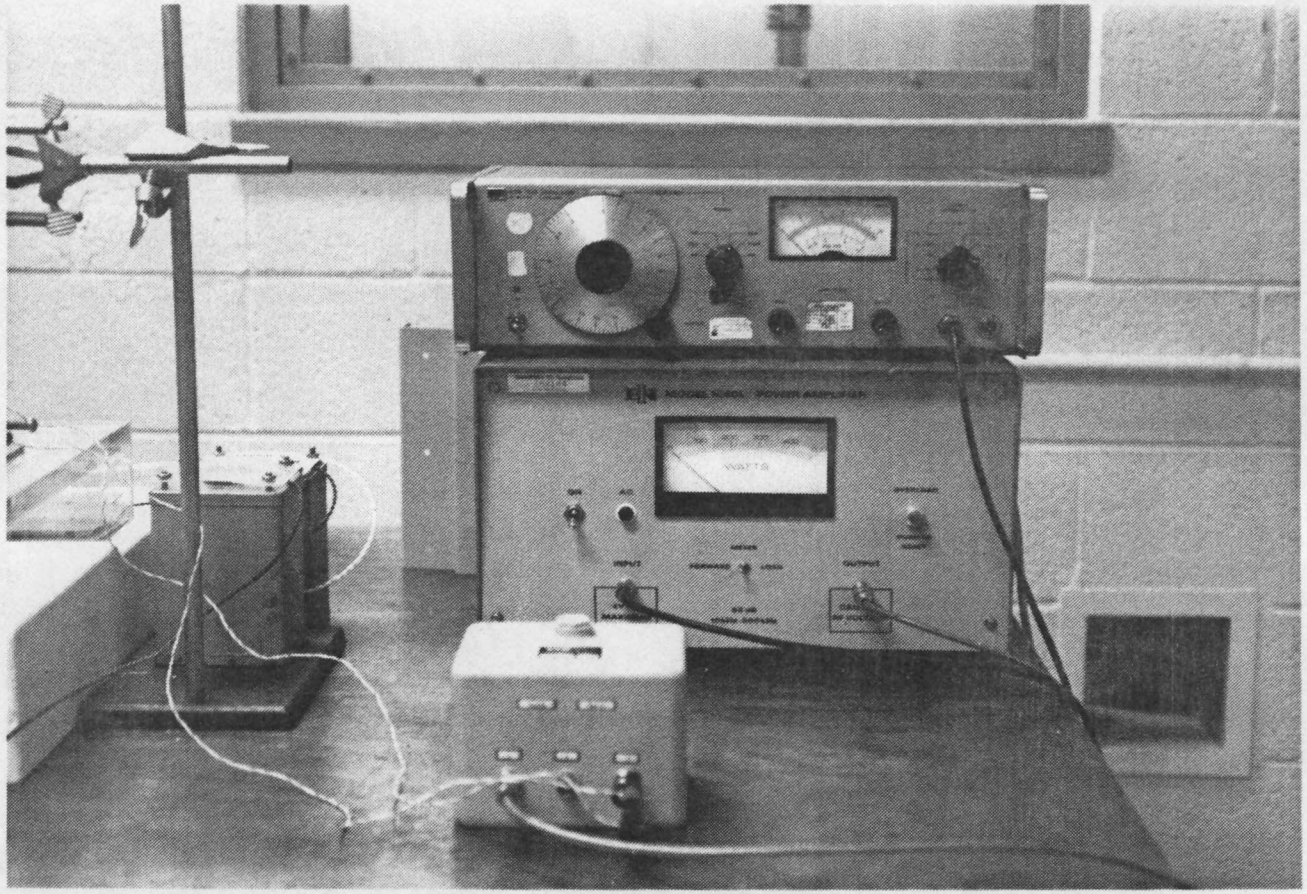

Figure 18.
A photograph of the levitator electronics

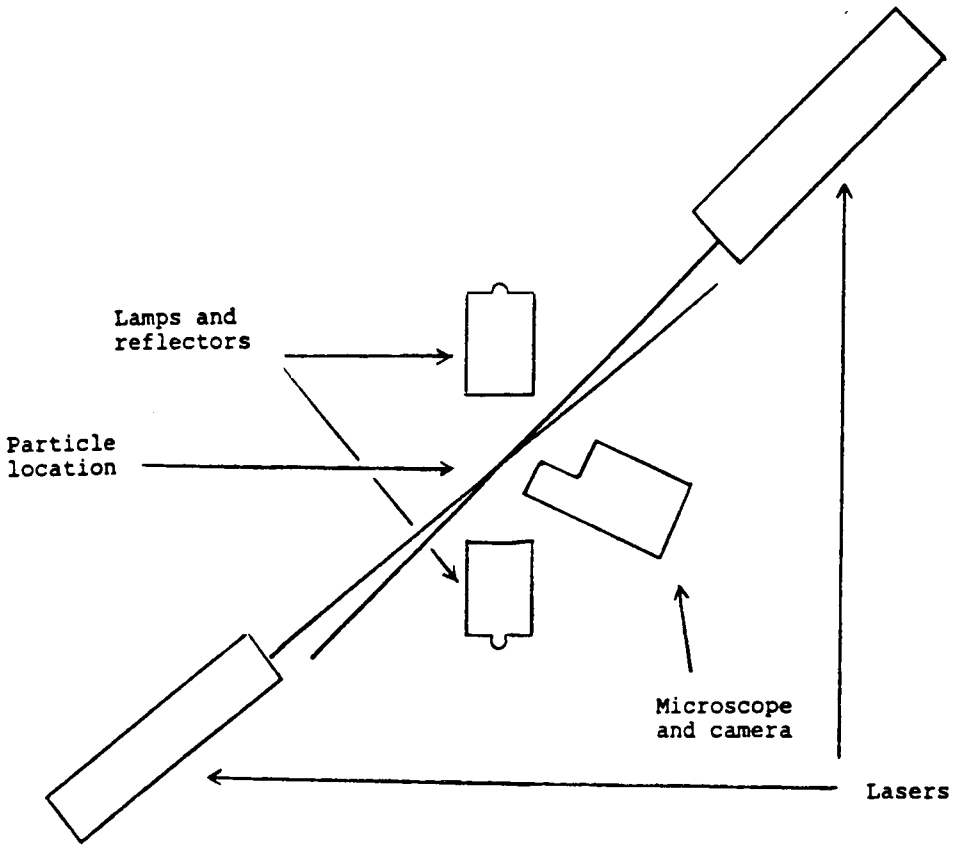

Figure 19.
The optical equipment

through radiation which a black, rough coal particle body absorbs well. The current and voltage of the lamps were matched to a standard outlet by running electrical leads from the lamps through two Malmac step-down transformers, and into a Staco Energy Products variable autotransformer which regulated the power output of the lamps.

The energy from the lamps is focused on the particle using ellipsoidal reflectors, one for each lamp. The reflectors, manufactured by Melles Griot, are made of nickel substrate electrolytically coated with a rhodium film. The ellipsoidal shape allows the mirror to condense an omnidirectional light source to a secondary focal point (see Figure 20). With this configuration, the particle heating rate may be estimated at 4650 K/sec when the lamps are first switched on. When the particle has reached steady state, the final temperature is estimated to be 1800 K (see the Appendix).

The filament of the lamp must be positioned at the precise point so that the filament`s image is on the particle. The broad-band assembly is held stationary by mountings made of plexiglass, rods and clamps. A convenient by-product of the lamps is that they provide the necessary illumination of the particle for high-speed microphotography.

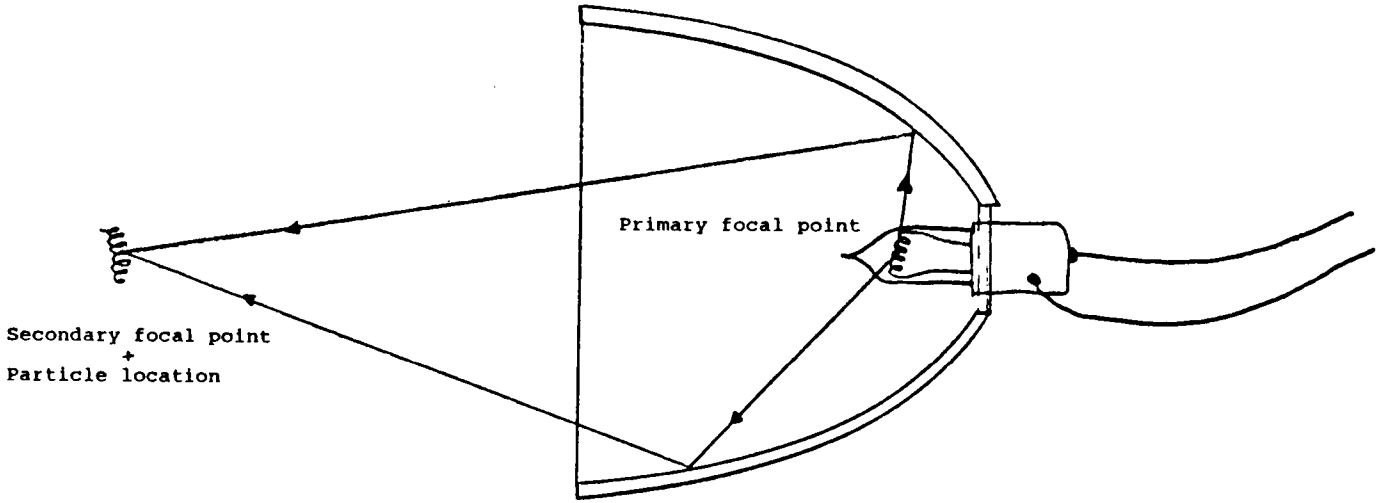

Figure 20.
The ellipsoidal reflector

Observation of submillimeter particles of coal samples during heating requires magnification to reveal the detail of external restructuring. A Nikon stereoscopic microscope was oriented horizontally on its heavy table stand to fit the particle in its viewing field while not obstructing the operation of the levitator. Either an observer can use the binocular lenses or a camera can be attached to the photo tube to take pictures. Because focal lengths are different, these two operations cannot be done at the same time. For a 35-mm camera, the coupling was obtained by a T-adapter on the camera, fastened to an aluminum tube which fits in the photo tube. Still photographs were taken in the early stages of the experiment to provide information about lighting, exposure times, shutter speeds and focusing.

A Hycam 16-mm movie camera was used for the high-speed photography. It was coupled to the microscope using an approach similar to that used for the 35-mm camera, but instead of being fastened to the microscope, the Hycam rested on the table and against the photo tube. By moving the microscope and the camera independently, risk of injury to either piece of equipment was minimized. Although possessing the capability to run at 11,000 frames/sec, the Hycam was operated between 500 to 1000 frames/sec which was adequate to arrest on film frames the

erratic motion of the particle in the levitator. Camera speed was limited by available light since the light intensity is roughly proportional to the inverse of magnification.

Equipment Testing

To fully prepare for taking high-speed pictures of coal particles undergoing pyrolysis in air, preliminary tests were performed on the acoustic levitator and the optical equipment. Specimens of various materials were introduced into the sound field to note their movement and stability. Single captive coal particles were heated with the lamps and lasers at the assumed levitating position. The experimental components were tested simultaneously as samples of styrofoam and wax were heat processed in the levitator and photographed with the 35-mm camera. Each stage of testing helped to build confidence that useful final results would be obtained.

Particles of different substances were levitated to study their behavior, to master the operation of the levitator and to test its limits. Styrofoam was suspended easily because of its low density. By adjustments to the driver-relector separation distance, a piece of styrofoam could be induced to spin on a vertical axis or remain essentially immobile. Figure 21 is a photograph of spinning styrofoam taken at 1/1000 sec. Wax, rubber and

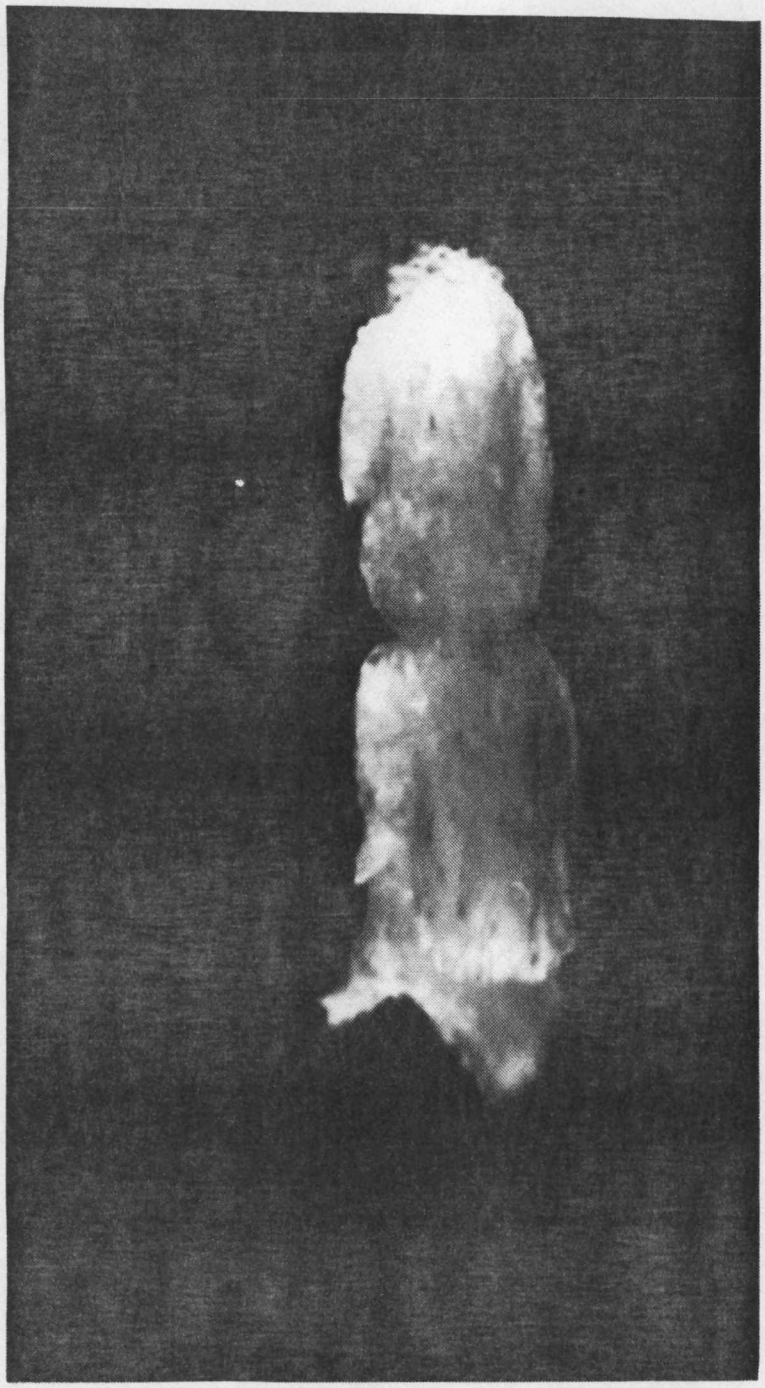

Figure 21.
A photogragh of spinning styrofoam

coal all exhibited the tendency to spin. Rotation of a particle on an axis was not considered a drawback but would actually help to distribute the temperature of a particle uniformly during heating. Micro-adjustments of the acoustic reflector were sometimes necessary to stop the particle from revolving in a circle or from swaying side to side. A piece of solder (a lead and tin alloy) was the heaviest sample levitated. It sometimes bounced up and down unstably in the acoustic field.

An OHIO no. 6 seam coal from Sunnyhill Mine was used for this research. Its ultimate and proximate analyses are given in Table 2. Preparation of the coal sample for heating required two operations. The coal was crushed and sieved into three size ranges (μm): from 300 to 355; from 355 to 420; and from 420 to 500. The particles were then placed into airtight jars to prevent preoxidation. The largest particles were used because they were physically easier to handle than the smaller particles and because particles up to 400 μm can be considered to heat uniformly (Smoot and Smith, 1985).

To test the optical equipment and to verify the coal as caking, single captive particles were heated with the focused light. One at a time particles were perched on thin wire. Microphotographs were taken before, during and after heating and are presented in Figures 22 to 25. The

Table 2.

Analysis of OHIO No. 6 Coal

Analysis (Dry Basis), Wt. %	
Ultimate	
C	59.8
H	4.3
N	1.1
Cl	0.006
S	3.51
Ash	23.68
O	7.55
Proximate	
Volatiles	34.76
Fixed Carbon	41.56
(Moisture)	(8.8)
Heating Value, Btu/lb	10,709.
CaO in Coal, Wt. %	0.31
Ca/S Mole Ratio	0.05
Sulfur in Ash (SO_3), Wt. %	1.1

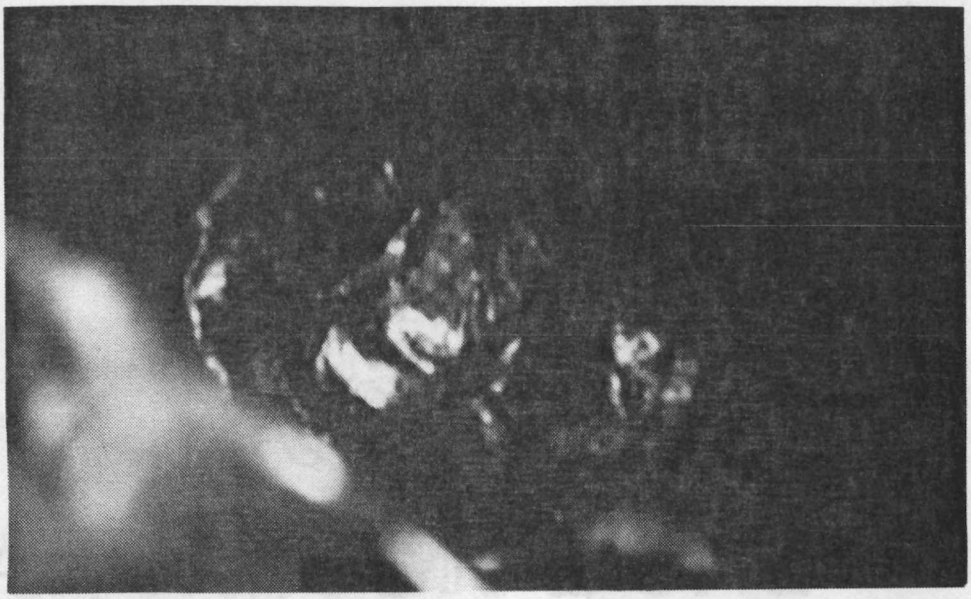

Figure 22.
A single captive coal particle

Figure 23.
The particle softening

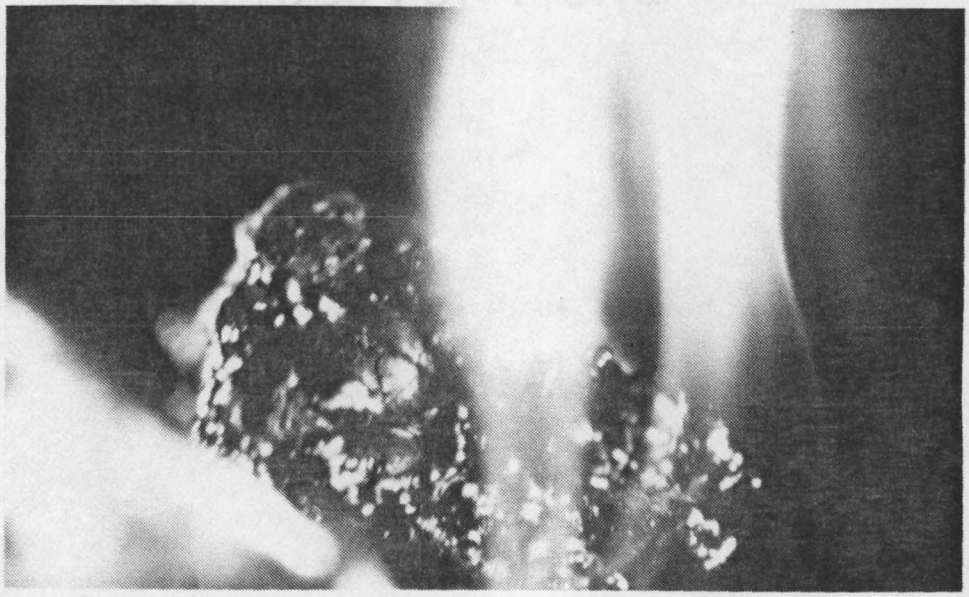

Figure 24.
Some volatile clouds

Figure 25.
The particle remnant

angular structure of the coal smoothed out as the coal softened and swelled. Clouds of volatiles, ejected as a result of decomposition, were sometimes dense enough to block the view of the coal. The remnant of the particle had many holes and cracks which the gases formed to make a path to exit the particle. Thus, the optical equipment was tested and the coal was verified as caking.

Particles of candle wax were levitated and irradiated to get a combined study of levitator stability and the optical equipment. Processing wax was found to be very routine. A piece of wax is lighter in weight than a similar sized piece of coal. It has a low melting point at about 70 °C. Figure 26 is a photo of an irregular piece of wax whose spinning motion was stopped by a short exposure time. As it turned to liquid (see Figure 27), it became spherical and was much more stable in the acoustic well. After the particle cooled, it's shape resembled a thick disk with rounded sides (see Figure 28). The symmetry was so precise that there was no observation of any spinning motion. Levitation of a solid and a liquid and a phase change was demonstrated. Observation of coal particle softening could be expected.

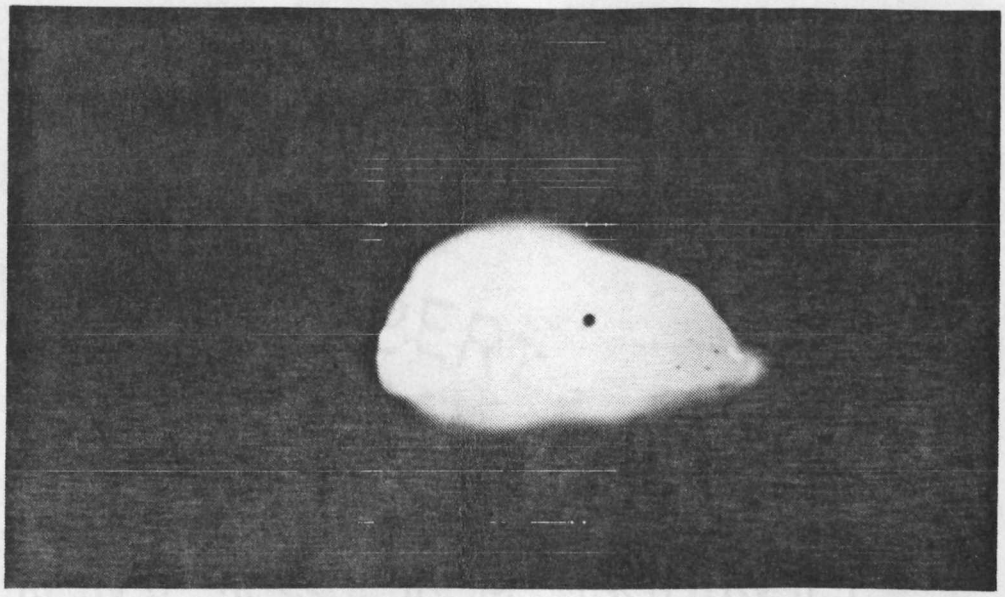

Figure 26.
A 1 mm sample of spinning wax

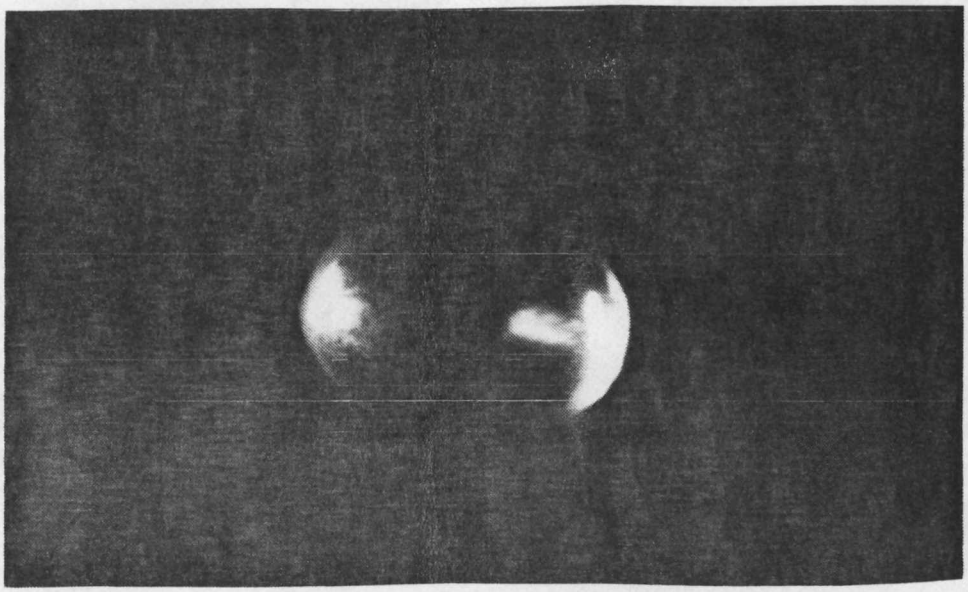

Figure 27.
Liquid wax

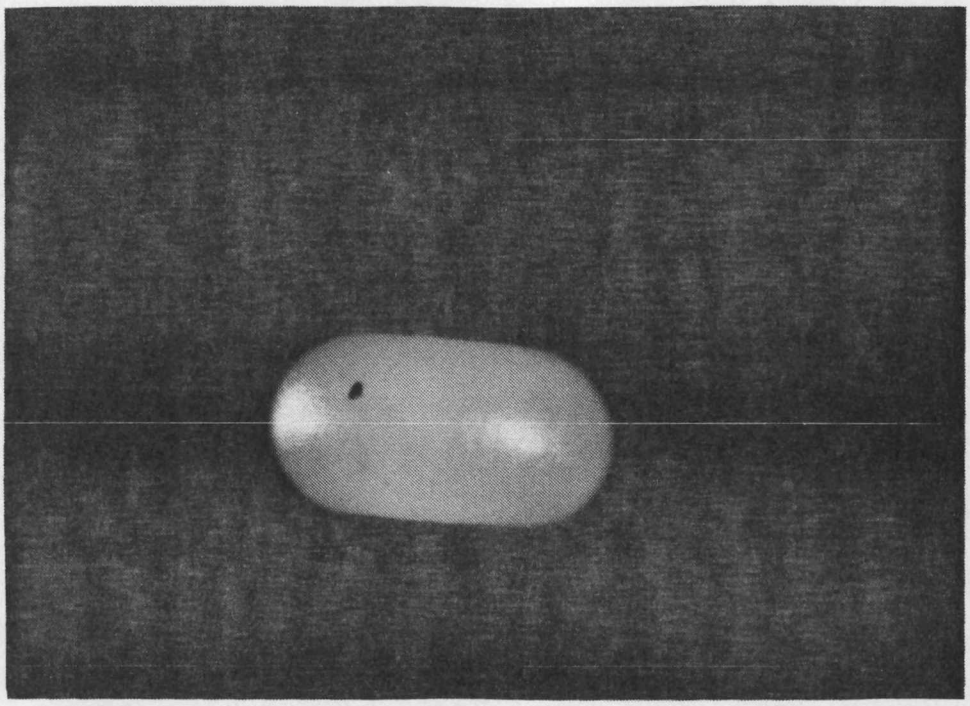

Figure 28.
Cool, reformed wax

CHAPTER IV

RESULTS AND CONCLUSIONS

Chapter IV discusses the information obtained pertaining to coal pyrolysis using the method described in Chapter III. In this chapter, microscopic observations are explained in detail. Some photographs are included and discussed. Finally, conclusions of this research effort are presented emphasizing the various successes and failures.

Results

Over the course of this investigation, over 1000 coal particles in the levitator were observed by microscopic viewing. Particle behavior was surveyed with and without irradiation. Notes were made on particle type, power input to the levitator, driver-reflector spacing, and lamp settings.

Any particle was stable prior to heating if the driver-reflector separation distance was adjusted correctly and the power input was at least 30 W. A particle was urged into the sound field by allowing the sound pressure to lift it off a wire "rake" into the nearest well. Vertical or lateral instability was slight but spinning was unavoidable. Particle sizes or shapes did not seem have any bearing on their stability.

However if a particle was flat or had a flat side, it tended to align that side parallel to the driver. If it was very small (100-200 μm), handling and acoustic liftoffs were difficult. The consistency and stability of levitated "cold" coal particles were routine throughout the research, using the final version of the levitator.

Upon heating, a problem was encountered that severly limited the study. When the lamps were switched on, a particle's stability was lost. At low lamp settings, where the temperature was below coal softening temperature (as evidenced by the earlier single captive particle studies), a particle would start shaking back, forth and/or up, down. Soon after it would exit the sound field and consequently the field of vision.

It was difficult to pinpoint the reason for this instability. Figure 29 shows the correlation for changes in wavelength (at 41 kHz) as the air in the sound field is warmed. The curve shows that the wavelength at 1000°F is 167 percent of the wavelength at 80°F. However, any air temperature changes are likely to be localized in the area immediately surrounding the particle. Radiation does not significantly heat an transparent medium such as air. To increase stability, the driver-reflector spacing was lengthened as much as possible with cold particles to account for a small change in wavelength upon heating.

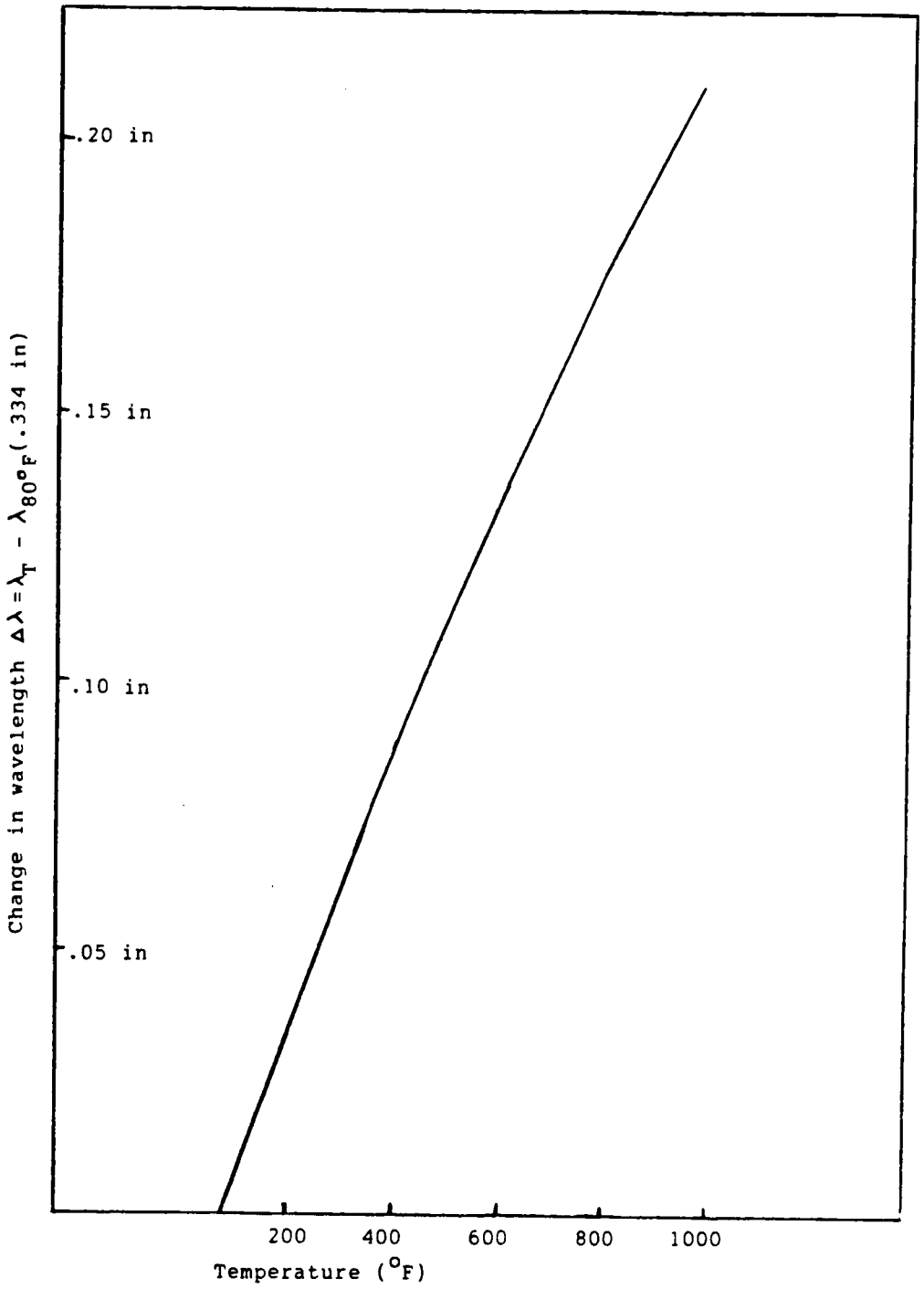

Figure 29.
The change in wavelength with temperature

Different particle shapes had different behavior with heating. Flat particles tended to stay in the well for the longest time. That they were the least heated is likely the reason. With the lamps oriented horizontally and the flat coal sample spinning horizontally, there was not much surface area to absorb the heat. The most interesting observations were of particles that were nearly spherical or symmetric in all directions. They were more stable (had less movement) than particles with asymmetric characteristics. Symmetry was presumed to be important for uniform heating.

The best observations were made when the lamps were switched on at their maximum setting onto a levitated particle. Figure 30 shows the most common occurence. As the lamps were switched on, the particle loses its stability and simply spins or rolls out of the field of vision. No obvious particle changes could be observed, possibly due to an absence of changes but probably due to the particle's behavior being too quick for the eye to follow.

Particles were seen exploding out of the well. If a particle stayed in the focal point of the lamps long enough (over one second) to absorb significant heat, its pressure would build up internally. This would occur from water rapidly vaporizing or higher temperature volatiles

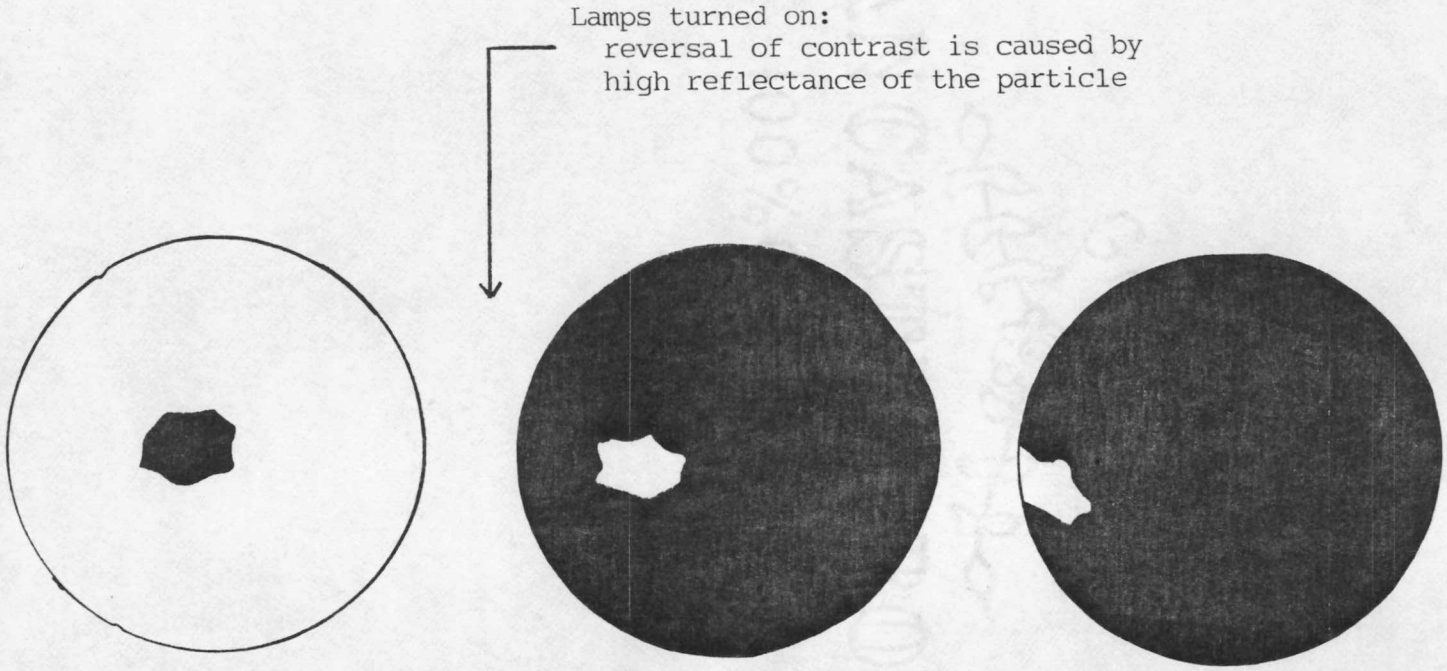

Figure 30.
A figure of a particle rolling out of the well

breaking away from the coal. The observed explosion (see Figure 31) only took a fraction of a second. The particle was so illuminated that points of light could be seen flying in different directions.

Occasionally, bursts of volatiles could be seen coming out of a particle. Instablility would accompany this jetting effect and most likely was partly caused by it. Figure 32 shows a sequence of events where the volatiles exit one side of the particle and propel the particle out of the sound field.

The spinning phenomenon was the greatest hindrance to accurate observations. Microscope focusing was estimated as closely as was practical to sharpen the outline of the blur. Coal softening could not be seen but swelling sometimes occurred as the overall size of the spinning particle appeared to increase. If volatiles were evolving from some particles, the spinning might be quickly fanning them out of sight.

The problems associated with personal observations of coal particle behavior in the levitator were amplified when trying to obtain photographic results. Figure 33 shows a typical photograph where the spinning was so great that the motion could not be stopped even at a shutter speed of 1/4000 second. Additionally, this picture is reasonably in focus, whereas in the bulk of the

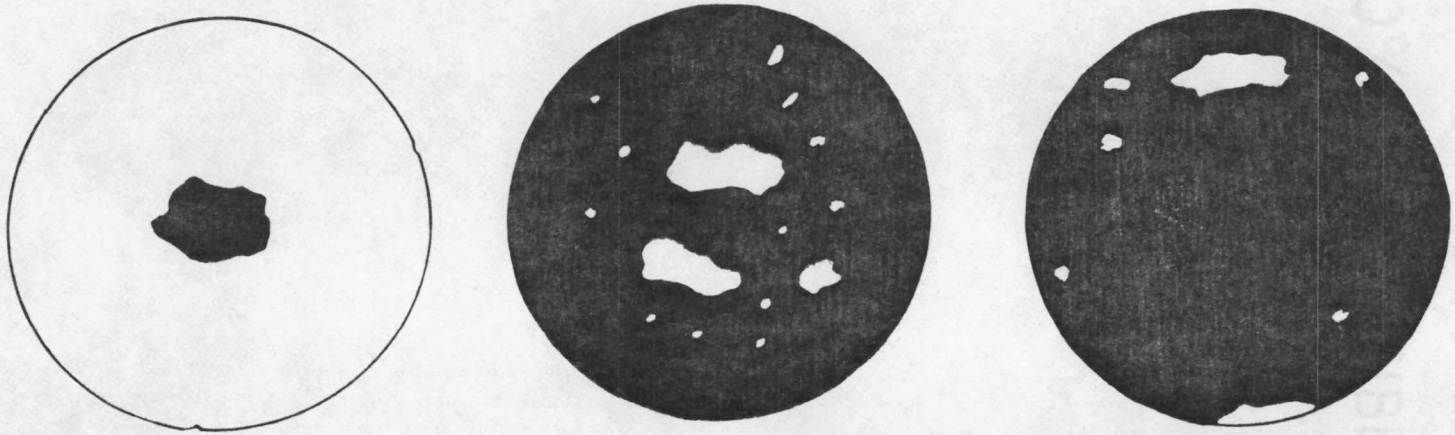

Figure 31.
A particle exploding out

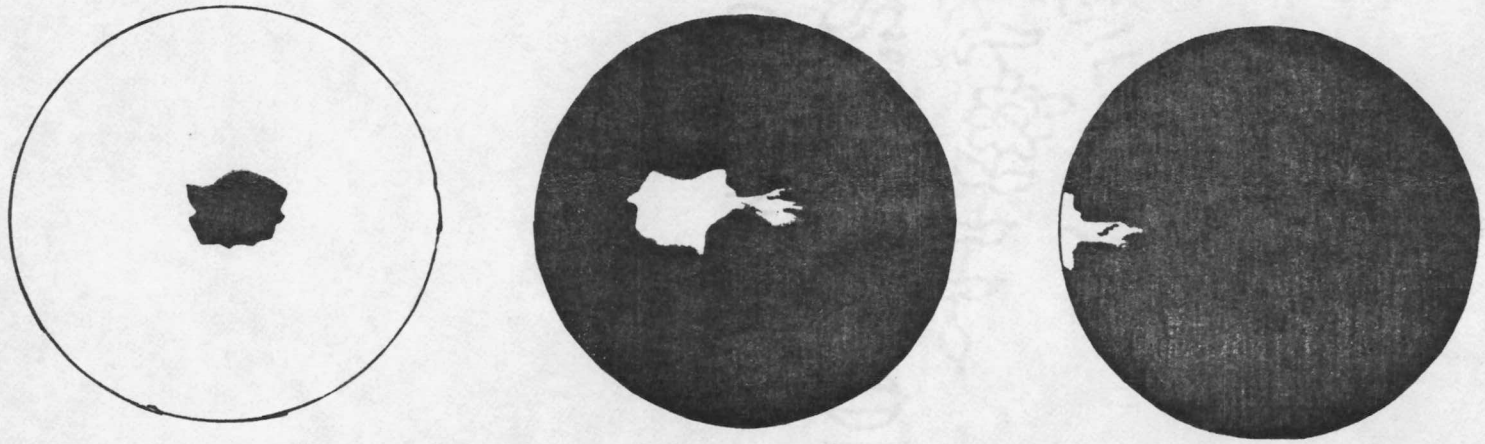

Figure 32.
A particle jetting out

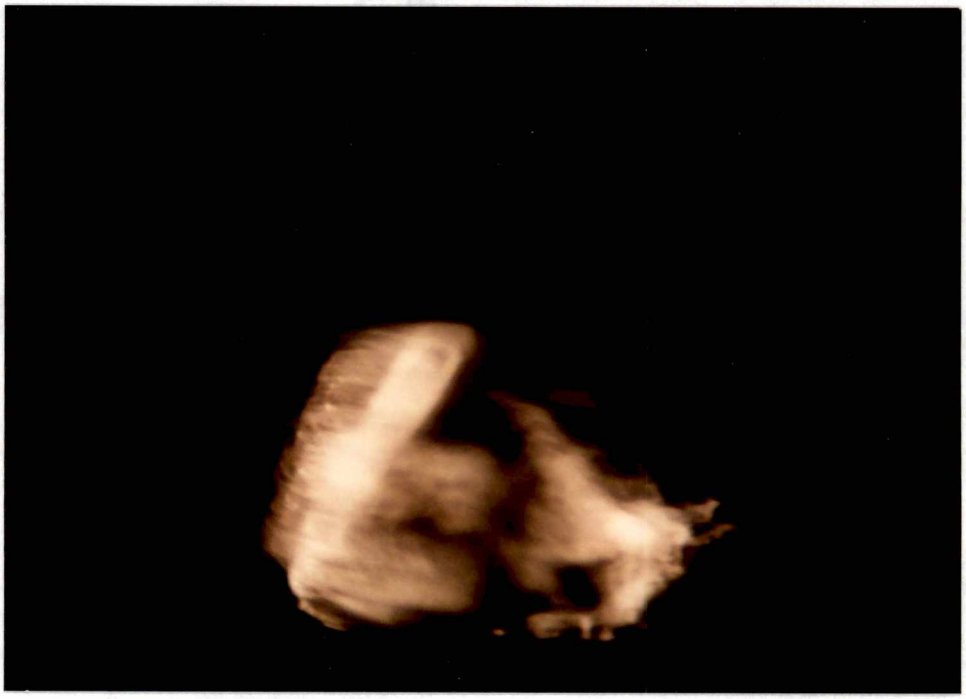

Figure 33.
A spinning coal particle

photographs taken, the particles moved out of the microscope's plane of focus leaving fuzzy blurs on the film. Compounding the problem is the fact that even the focused radiation was not bright enough to expose the film at higher shutter speeds. The approach required to obtain good quality photographs was to link together timing, quantity (a large number of attempts), and luck.

The following series of figures are reasonably good quality photographs of cold coal particles. They were taken as soon as the lamps were switched on. Figures 34, 35 and 36 show particles very roughly symmetrical in shape but angular and planar in texture similar to the captive particle before heating. A flat shaped particle is shown in Figure 37. It is reasonably accurate to conclude that their (Figures 34-37) spin was not as pronounced as a typical particle. Capturing the moderately spinning particles was another factor in getting decent pictures.

Figures 38 and 39 show an interesting variation in cold particle behavior. This particle is not only spinning on the vertical axis (shown by its change in orientation) but is rotating on a horizontal axis. These photographs were unique out of the entire set.

The next series of photographs are of particles that have been given some time to absorb the radiation. They

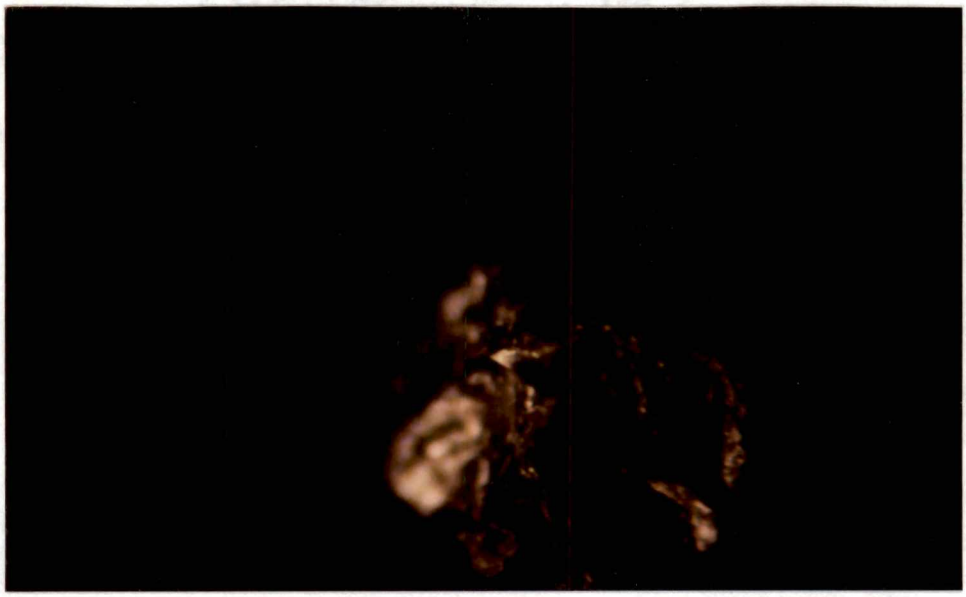

Figure 34.
A cold particle1

Figure 35.
A cold particle2

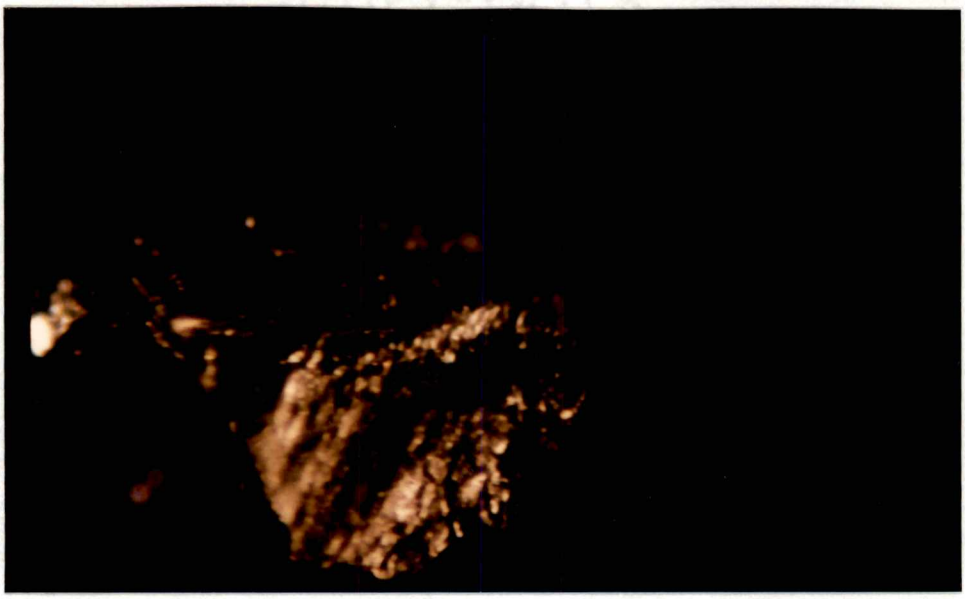

Figure 36.
A cold particle3

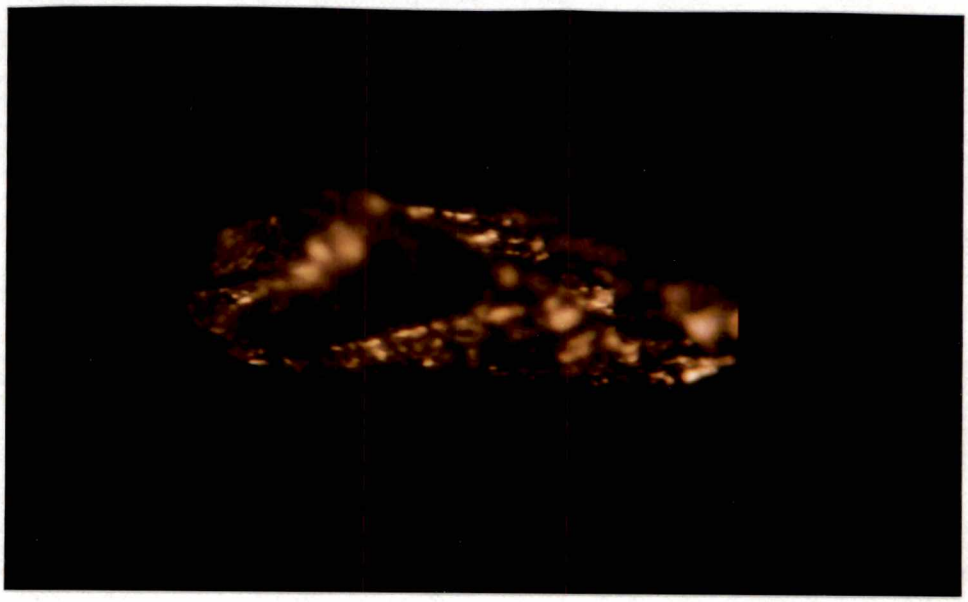

Figure 37.
A flat particle

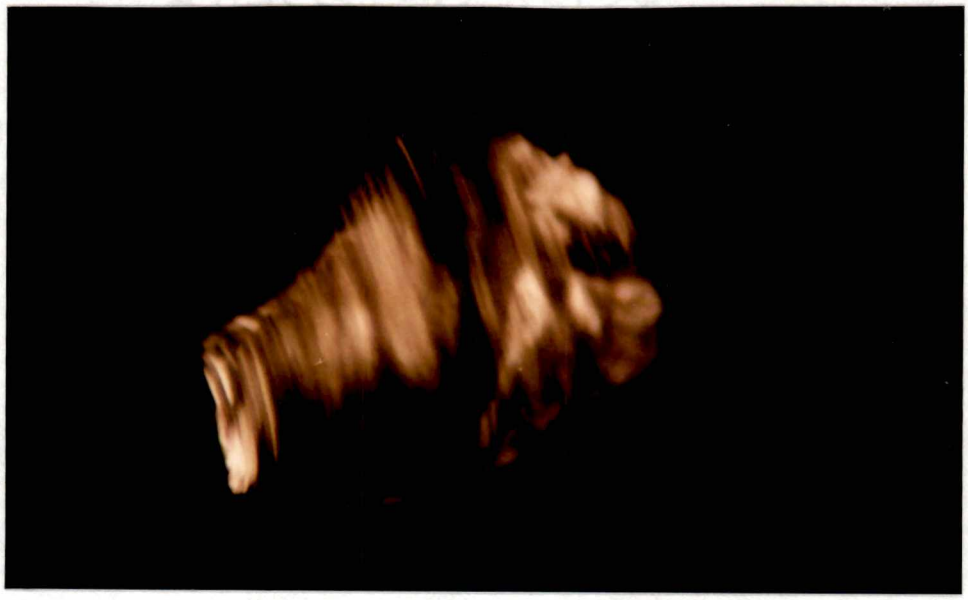

Figure 38.
A particle with an unusual spin

Figure 39.
The same particle with an unusual spin

are the accumulated set of pictures which were lucky enough to be shot right before the particles exited the acoustic well. (Many frames of the negatives were completely blank because the camera operator missed that precise moment.)

Figures 40-42 show particle bubbling occurring as the volatiles decompose from the coal molecules and induce softening. The softening appears to be spatially uniform. The restructuring process of these levitated samples is is very comparable to the captive sample particle. Possibly due to their size (approximately 500 μm), the particles' overall shape does not seem to have changed much. Instead of becoming spheres, small bubbles appear over their whole surface. If the particles had remained stable in the well, the gases would have probably broken out of the bubbles, forming microjets (which have been occasionally observed) and leaving blowholes in the char.

Figure 43 is an example of non-uniform softening or the onset of softening. As one can see at the right side of the particle, at the top and bottom corners, bubbles are forming. The rest of the particle had not absorbed enough heat to begin softening.

The last photograph in this series requires some speculation. Shown in Figure 44, this particle's external texture is extremely rough appearing like sandpaper.

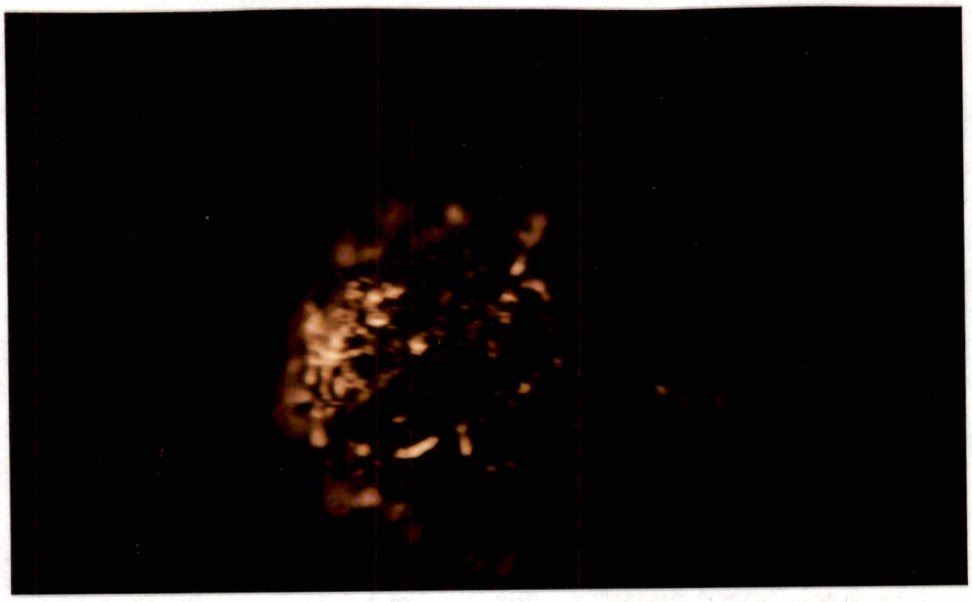

Figure 40.
A particle bubbling1

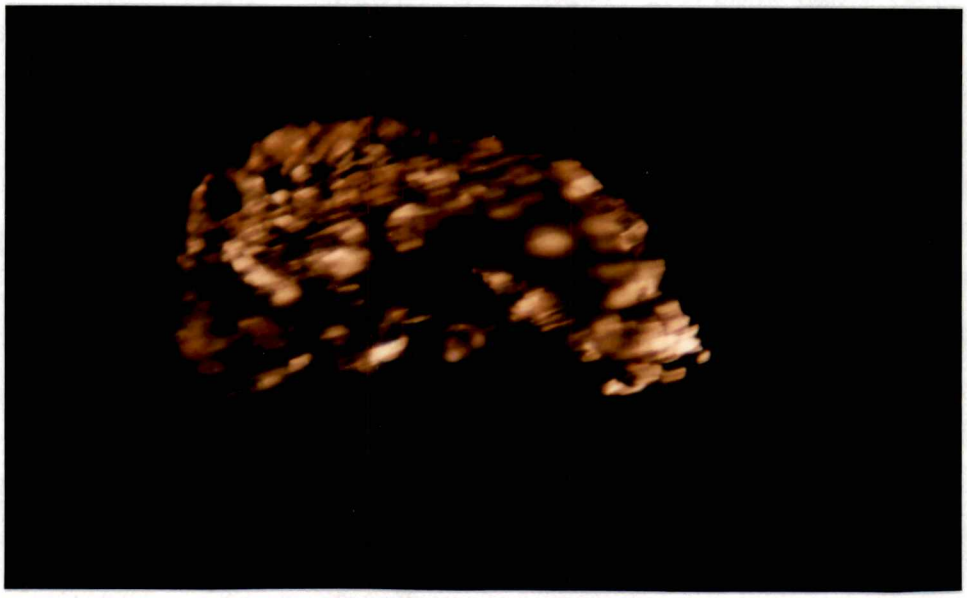

Figure 41.
A particle bubbling2

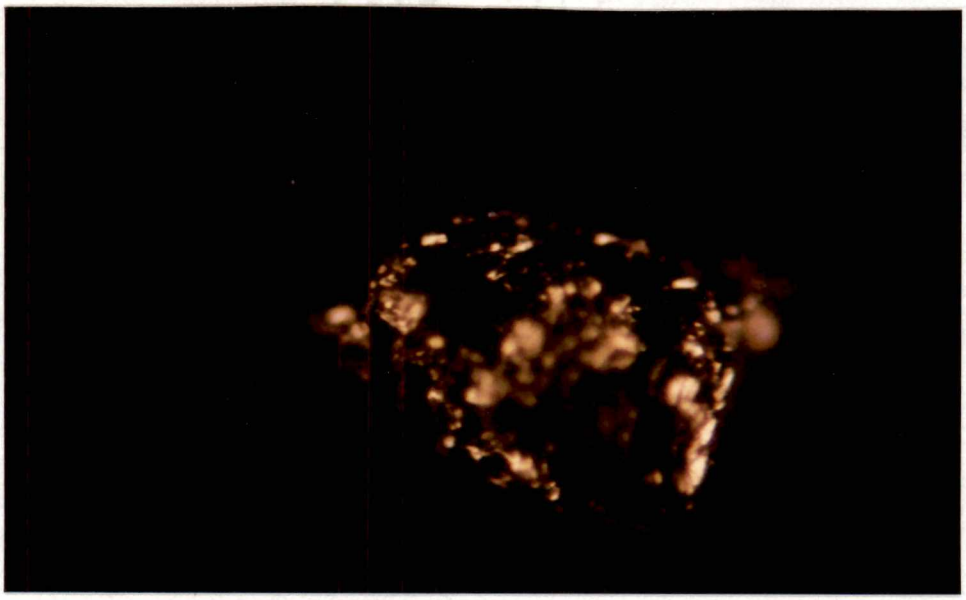

Figure 42.
A particle bubbling[3]

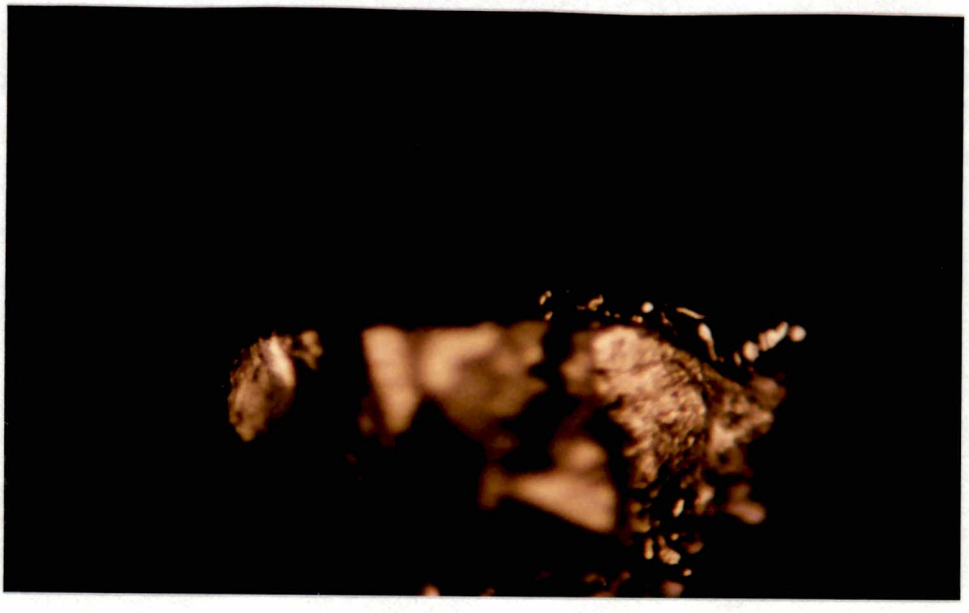

Figure 43.
A particle with non-uniform bubbling

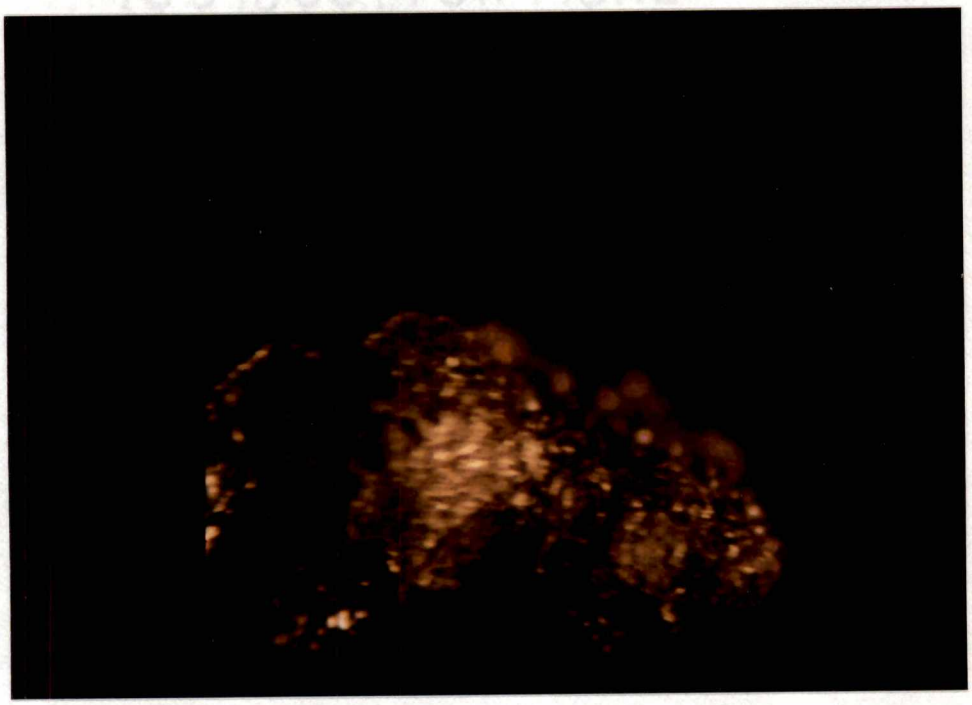

Figure 44.
A particle with a rough texture

This particle's structure had surely changed because all the particles were observed cold before being inserted in the levitator and heated, and they all displayed the same angular characteristics. Because of its apparent difference to the previous particles, and to account for its granular texture, it might be speculated that either it has a high mineral content or that it has somewhat oxidized.

A particle rolling out of the well was filmed with the high speed movie camera. In the sequence of photographs (see Figure 45) the observation of Figure 30 (page 71) is verified. Since this phenomenon was the most common, it was logical to expect that simple particle roll out would be filmed almost every time.

Conclusions

The positioning of various cold materials, including coal, by acoustic levitation was demonstrated. Then, the melting of wax was practiced routinely. Because of these successes, the instability of heated coal was not anticipated. It was a problem that was dealt with by providing conditions that would keep a particle in position for as long as possible. The driver-reflector spacing was increased to account for wavelength changes with temperature. Nearly spherically shaped particles were used.

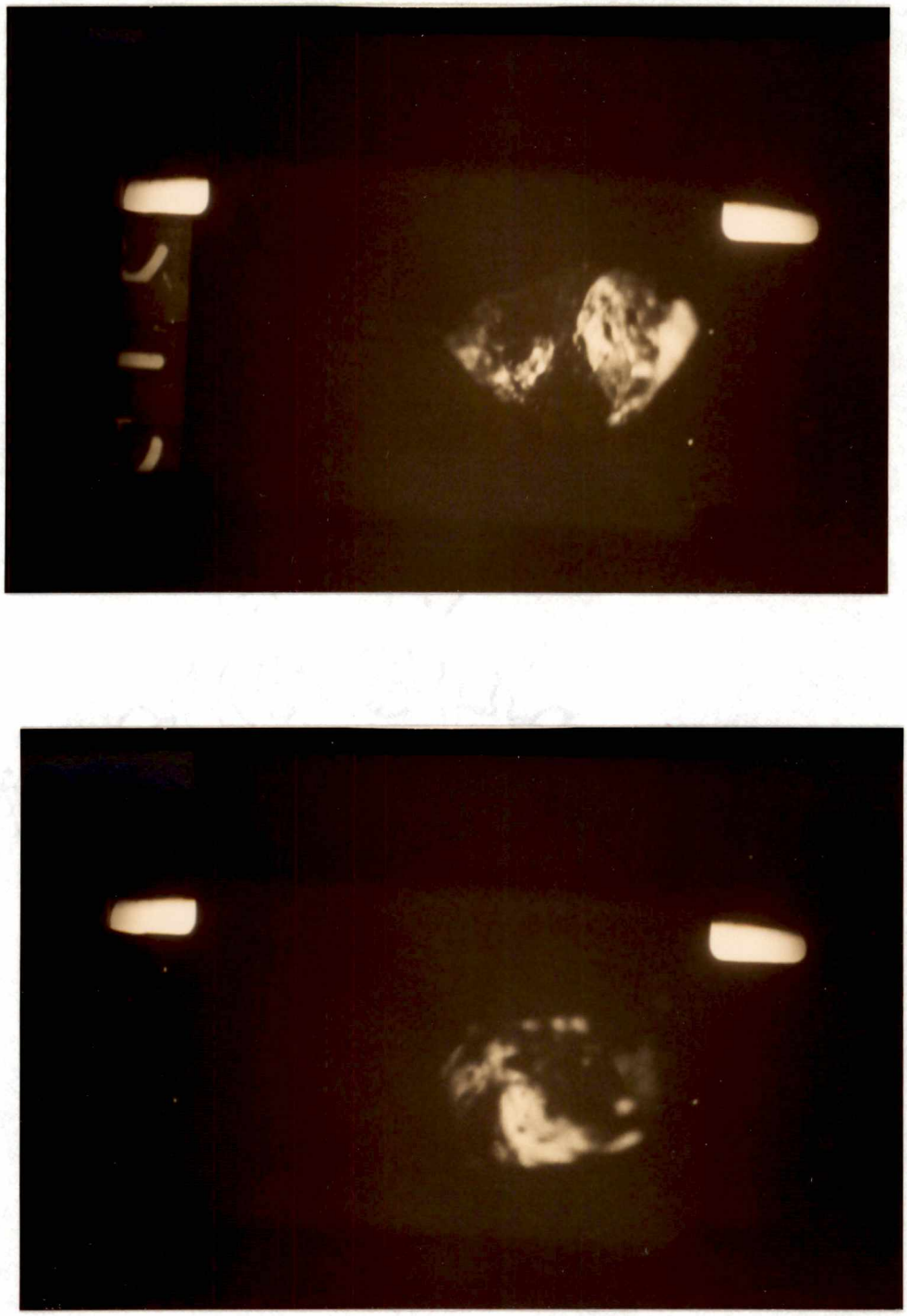

Figure 45.
Some photographs from the Hycam film

Figure 45. (con't)
Some photographs from the Hycam film

The results from observed particles demonstrated that some pyrolysis took place during heating. With the coal being solid body contact free, some coal behavior was seen that was not seen with captive particles. It was personally observed how internal pressure build up could be released. Particles would either explode and break apart into small pieces or microjets would form, propelling the particles through the air.

Quality photography did not become routine. Because particle positioning was not as stationary as desired upon heating, much of the technique was dictated by chance. However, the coal softening figures are a bit of proof verifying the microscopic observations. Since in caking coals, softening precedes volatile evolution, explosions or microjets are the next steps driving the particles out of the well. The particles tended to soften uniformly. In a captive sample radiative heating experiment, a particle's caking would be inhibited in any area of solid body contact. This was observed during the filming of Figures 22-25 (pages 62,63). The area of coal-wire contact was the last to soften.

CHAPTER V

SUMMARY

An acoustic levitator was designed and constructed to hold coal particles stationary in air. It consisted of a simple, single axis driver-reflector system that allowed for optical focusing, observation and photography as described in Chapter III. This research demonstrated the development of an acoustic levitation apparatus to form a potentially improved coal pyrolysis experiment over previous experiments with their drawbacks discussed earlier. The main advantages of this study were being able to clearly view a known particle reacting because only single particles were used and holding that particle in position without any solid body contact when heating.

The literature review of Chapter II and the equipment testing section of Chapter III helped to provide expected results from the experiment. From the survey of the physical description of pyrolysis and from the single captive particle study, the behavior of coal upon heating was anticipated. The results discussed in the previous chapter on coal swelling and softening, volatile jetting and particle shattering verified the documentation of previous studies. The fact that the results of Chapter IV were obtained in a more realistic environment (suspension

in a gas, radiative heating) displays the progress that this technique can contribute to understanding pyrolysis.

In light of the problems discussed in Chapter IV, improvements on the technique can be suggested. The stability problem must be eliminated to observe the pyrolysis process completely through the temperature stages: from water evolution, through active thermal decomposition, to the formation of the char particle. To accomplish this, the hemispherical focusing dish might be used to add a greater symmetrical restoring force. Or a triple-axis system could be utilized which would add additional horizontal restoring forces and should stop particle spinning, (the other problem discussed, but of relatively minor importance). The disadvantage of these systems is that they require long range optical equipment that would not interfere with the levitator or the sound field.

Should the instability drawback be removed from the technique, important potential additional studies may be undertaken. They are listed as follows:

1. the apparatus could be sealed into an inert gas environment to prevent any oxidation,
2. mass loss could be measured,
3. mass loss as a function of time-temperature correlations could be determined using

a. a multi-color pyrometer or

b. FT-IR spectroscopy,

4. this data could be used to develop a pyrolysis model or extend an existing model to high temperatures (T>2000 K),
5. to determine volatile composition, the pyrolysis products could be admitted into an analyzer such as

   a. a gas chromatograph or

   b. a mass spectrometer,

6. heating time in air could be increased to study volatile and char combustion, and obtain burning rates, mechanisms, etc.

The complete understanding of pyrolysis is important for the future when our limited supply of petroleum is almost exhausted and our need for an efficient coal conversion technology becomes apparent.

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APPENDIX

APPENDIX

List of Symbols

Symbol	Definition	Units
A	area	[m]
a	constant	[sec^{-1}]
b	constant	[K/sec]
Bi	Biot number	
c	specific heat	[J/kg•K]
D	diameter	[m]
g	gravitational acceleration	[m/sec^2]
$\bar{h}$	average heat transfer coefficient	[$W/m^2 \cdot K$]
k_a	thermal conductivity of air	[W/m•K]
k_c	thermal conductivity of coal	[W/m•K]
Nu	Nusselt number	
Ra	Rayleigh number	
r	radius	[m]
T_c	coal temperature	[K]
T_a	ambient temperature	[K]
T_l	lamp temperature	[K]
t	time	[sec]
V	volume	[m^3]
α	thermal diffusivity	[m^2/sec]
β	volumetric thermal expansion coefficient	[K^{-1}]
η	reflector efficiency	

ν	kinematic viscosity	[m^2/sec]
ρ	coal density	[kg/m^3]
σ	Boltzmann's constant	[$W/m^2 \cdot K^4$]
θ	temperature variable	[K]

Biot Number Calculation

In order to assume a 500 μm diameter particle spatially lumped,

$$Bi = \frac{hr}{3k_c} < .1$$

$r = 2.5 \times 10^{-4}$ m
$k_c = .26$ W/m•K

solving for h,

$h < 3120$ W/m²•K

Heating Rate Calculation at Start Up

An estimate of the average heat transfer coefficient is as follows (free convection correlation referenced from Incropera and DeWitt, 1985):

$$\bar{h} = \frac{k_a}{D}\overline{Nu}$$

$$\overline{Nu} = 2 + .43(\overline{Ra})^{.25}$$

$$Ra = \frac{g\beta\Delta T D^3}{\nu a}$$

$$\overline{Ra} = \frac{Ra(\Delta T = 1700\ K) + Ra(\Delta T = 100\ K)}{2}$$

$g = 9.8$ m/sec²
$\beta = 2.725 \times 10^{-3}$ K^{-1}
$D = 5 \times 10^{-4}$ m
$\nu = 1.589 \times 10^{-5}$ m²/sec
$a = 2.25 \times 10^{-5}$ m²/sec
$k_a = .0263$ W/m•K
$T = 300$ K
$T = 2000$ K

$\overline{Ra} = 8.41$

$\overline{Nu} = 2.73$

$\overline{h} = 144\ W/m^2 \cdot K$

The heat balance equation is,

$$\rho cV\frac{dT_c}{dt} = \eta\sigma A(T_l^4 - T_c^4) - hA(T_c - T_a)$$

$\theta = T_c - T_a$ (T_c^4 comparatively small)

$$\rho cV\frac{d\theta}{dt} = \eta\sigma AT_l^4 - hA\theta$$

$\rho = 1300\ kg/m^3$
$c = 1260\ J/kg \cdot K$
$V = 6.54 \times 10^{-11}\ m^3$
$\eta = .7$
$\sigma = 5.67 \times 10^{-8}\ W/m^2 \cdot K^4$
$A = 7.85 \times 10^{-7}\ m^2$
$T_l = 2000\ K$
$T_a = 300\ K$

$$\frac{d\theta}{dt} + a\theta = b$$

where $a = \frac{\overline{h}A}{\rho cV} = 1.055\ sec^{-1}$

$$b = \frac{\eta\sigma AT_l^4}{\rho cV} = 4650\ K/sec$$

solving and applying the initial condition, $\theta(0) = 0$ gives

$$\theta = \frac{b}{a}(1 - e^{-at})$$

differentiating and using $\theta = T_c - T_a$ gives

$$\frac{dT}{dt} = be^{-at} \quad \text{at } t = 0, \quad \underline{\frac{dT}{dt} = 4650\ K/sec.}$$

The Final Temperature

The steady state heat balance equation is from [6], $dT/dt = 0$,

$$\eta\sigma(T_l^4 - T_c^4) = h(T_c - T_a)$$

iterating for T_c gives $\underline{T_c \text{ final} = 1800\ K.}$

VITA

Joseph E. Marquis was born in East Providence, Rhode Island on April 10, 1960. He attended North Kingstown High School and graduated in June, 1978. The following August he entered Clarkson College in Potsdam, New York. In May, 1982, he received a Bachelor of Science degree in Mechanical and Industrial Engineering.

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